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## Radiological assessment of past, present and potential sources to environmental contamination in the Southern Urals and strategies for remedial measures (SUCON)

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# **Radiological Assessment of Past, Present and Potential Sources to Environmental Contamination in the Southern Urals and Strategies for Remedial Measures (SUCON)**

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**Abstract** This report summarises work done on the SUCON Project during 1996-1999 (European Commission Contract No. FI4C-CT95-0001). The project has focused on three major objectives: 1) An assessment of the radiological consequences of the contamination of the South Urals and the Ob river system from the production of plutonium at "Mayak", 2) The development of models to calculate doses to individuals and populations in the South Urals using environmental data, and 3) The intercomparison, harmonisation and standardisation of techniques used in dose reconstruction and specification of good practice in particular with regard to remedial measures.

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# 1 Introduction

Western laboratories became involved in the studies of the radioactive contamination of the Southern-Urals, when a delegation from the IUR (supported by the Radiation Programme of the CEC) visited the Southern Urals in 1990. A co-operation between the Institute of Plant and Animal Ecology (IPAE), Ekaterinburg (RU), the Institute of Biology of the Southern Seas (IBSS), Sevastopol (UA) and Risø National Laboratory (RISØ), (DK) was established. In 1994 INTAS selected a joint project (INTAS-94-1221) on radioecological studies in the Southern Urals for financial support (70 KECU). The participants were RISØ (DK), NRPB (GB), IPAE (RU) and IBSS (UA). INTAS projects only cover coordination costs for EU-participants, hence other sources of financial support are requested. The Danish Ministry of Interior have for the period 1994-98 supported the project with approximately 0.4 MECU.

Within the Joint Russian-Norwegian collaboration on "Possible impact of the MAYAK PA activities on the radioactive contamination of the Barents and Kara Seas, 3 Norwegian Institutes; Norwegian Radiation Protection Authority (NRPA), Agricultural University of Norway (AUN) and Institute of Energy Technology (IFE) collaborate with MINATOM, Moscow (RU), Typhoon, Obninsk (RU) and MAYAK PA, Chelyabinsk-65 (RU). NRPA co-ordinates the Norwegian work while MINATOM is co-ordinating the work on the Russian side. The project is part of the joint program under the Russian - Norwegian Environmental Commission and is supported by the Russian and the Norwegian Governments.

MAYAK is located at 55°44'N 60°54'E 70 km north of Cheliabinsk and 15 km east of the city of Kyshtym in the southern Urals. MAYAK covers an area of 90 km<sup>2</sup> and was operated by the former Ministry of Nuclear Power of the USSR (Now Ministry of Atomic Energy of Russia).

The implementation of the nuclear programme in Cheliabinsk region in the Ural involved contamination of the environment with long-lived fission products resulting from both accident conditions and the routine operation of the utility, particularly, in the initial period i.e. the late forties and early fifties.

Three major events have contributed significantly to the contamination of the Urals.

- a. During 1949-1952 liquid medium & low-level radioactive waste was disposed directly into the Techa river system (fig. 1).
- b. In Sept. 1957 a tank containing high-level rad-waste exploded at Kyshtym and contaminated the environment in a NNE direction.
- c. During 1967-1970 a windborn contamination of the Urals occurred due to dispersion of activity from the shores of Lake Karachay, which have been used for the deposition of high-level rad-waste.

Beside these major events the daily operation of MAYAK may have contaminated the environment by routine discharges.

At Ekaterinburg a fast breeder reactor (Beloyarsky, BN-600) is operated. Also this plant may have contributed radioactive contamination. Finally unknown sources should be kept in mind.

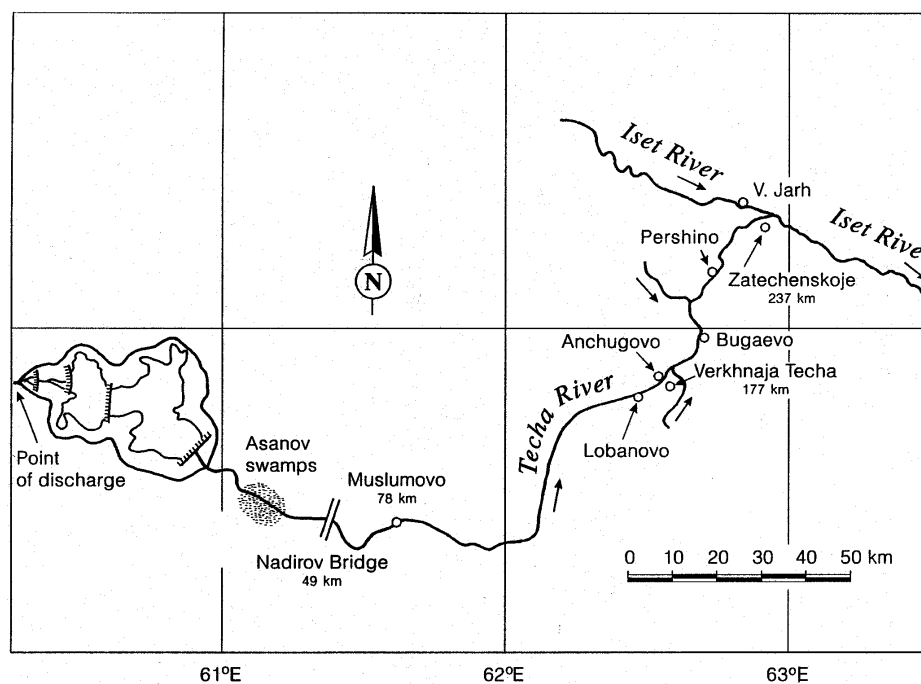


Figure 1. Techa river.

## 2 Objectives

The project focuses on three major objectives viz.

- a. An assessment of the consequences of the contamination of the South Urals and the Ob river system from the production of plutonium at “Majak”.
- b. The development of models to calculate doses to individuals and populations in the South Urals using environmental data.
- c. The intercomparison, harmonisation and standardisation of techniques used in dose reconstruction and specification of good practice in particular with regard to remedial measures.

More specific the objectives are

- To produce a joint Western-Russian report on the sources contributing to radioactive contamination of the “Majak” environment.
- To compare the inventories of radionuclides from Russian and western estimates based on environmental samples and measurements in the South Urals.
- To determine the exposure pathways to be considered in the dose assessment, together with the data required.
- To collect environmental measurements, habit and other relevant data through partners working in the Southern Urals region.
- To develop models where necessary for carrying out the dose assessment.
- To collect and measure teeth collected from the local population for levels of <sup>90</sup>Sr, Pu and Am.
- To consider strategies for the reduction of the radiation doses received by critical groups in the Southern Urals.
- To consider strategies for reclamation of contaminated land areas.

- To estimate the global impact, i.e. radionuclide inventories, of discharges to the Ob River system.
- To achieve new knowledge on the (long-term) behaviour of less well known radionuclides in the environment, in particular very long-lived radionuclides.
- To exchange information and samples from the various environmental activities carried out in the Urals by both European and American institutions in order to avoid unnecessary duplications and thus make the studies more cost effective.

## 3 Progress and Results

### 3.1 Review of Existing Knowledge

Since the early 1990s, confirmation of the nuclear activities at Mayak and releases of radionuclides to the environment have become available in a number of publications (e.g. Trapeznikov *et al.* (1993); JNREG (1997); Romanov (1995); Bradley & Jenquin (1995a); Malyshev *et al.* (1997)).

#### 3.1.1 Site development and description

The Mayak Production Association (PA) is located close to the town of Ozyorsk (Chelyabinsk-65) between the cities of Yekaterinburg (Sverdlovsk) and Chelyabinsk just east of the Ural mountains (Figure 1.1). The enterprise was established at the end of the 1940s to produce and reprocess weapons-grade plutonium. Site operations are currently under the jurisdiction of the Russian Federation's Ministry of Atomic Energy.

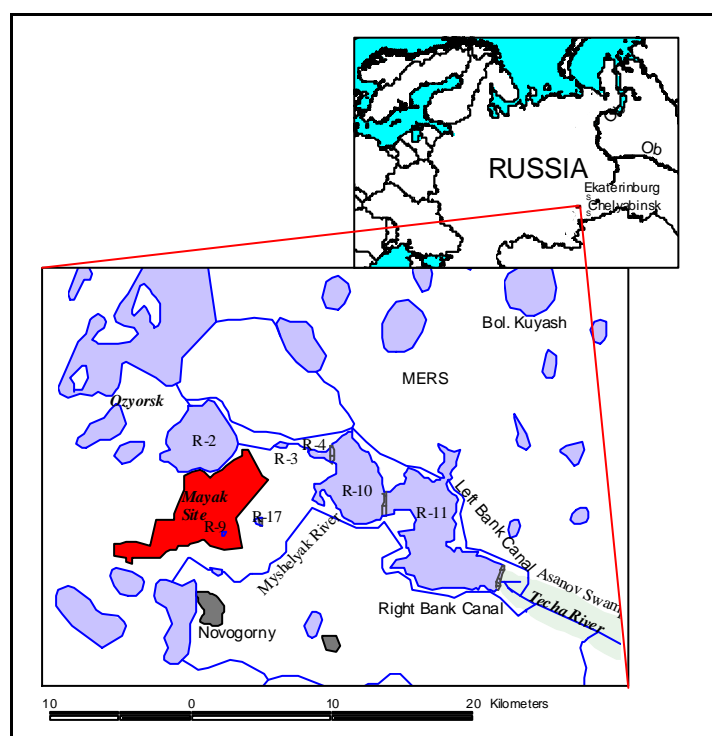


Figure 1.1. Map showing the location of Mayak PA.



Operation of the first military reactor began in June of 1948. At that time, other facilities were commissioned including a radiochemical plant for the separation of plutonium from uranium and fission products and a plant for the conversion of plutonium concentrate to high purity metallic components for atomic bomb production. Between 1949 and 1955 five nuclear reactors were built at the site and by 1979 a seventh reactor was in operation. In 1977, a new radiochemical plant of reprocessing nuclear fuel from civil reactors was completed. Since that time, Mayak has been reprocessing spent fuel from VVER-440 reactors, BN-350 and BN-600 fast breeder reactors, research reactors and nuclear vessel power units. Modernisation and expansion of the site also occurred in the 1970s. Production of weapons-grade plutonium was terminated in 1987 and of the original 7 military reactors, 5 uranium-graphite reactors were shut down between 1987 and 1991. The remaining 2 reactors currently produce radionuclides for military and civilian use. Today, the Mayak PA site also comprises of a radioactive isotope plant supplying radionuclides and radiation sources internationally, an instrument plant manufacturing measurement and automation equipment, a central laboratory and a experimental scientific research station.

A number of natural lakes and ponds on the Mayak site have been used as reservoirs for the management of intermediate and low-level radioactive effluent. These include Lake Karachay (Reservoir 9) and Lake Kyzyltash (Reservoir 2), the artificial ponds (Reservoirs 3, 4 and 17) and the artificial reservoirs created by damming the Techa River (Reservoirs 10 and 11) as shown in Figure 1.1. By-pass channels at the head of the Techa River were also built in order to reduce the amount of contamination entering the main river. Nevertheless, high levels of radioactive contamination in the area have made it necessary to establish a «Health Protection Zone» covering an area of approximately 350 km<sup>2</sup> around the site. Both agricultural practices and permanent residence are forbidden within this zone, although the area is still used for research purposes. A larger area has also been designated as a «Observation Zone» where regular monitoring occurs but where agricultural practices are permitted.

### 3.1.2 Radioactive waste management

Approximately 30 000 PBq of liquid and solid radioactive waste, including waste contained in the industrial reservoirs, has been accumulated at Mayak, according to the results of an inventory assessment made in 1990. High-level wastes (HLW) form a significant fraction of the total waste activity. Of the 23 500 PBq classified as HLW in March 1995, approximately 8 700 PBq was in the form of vitrified waste and approximately 14 800 PBq in the form of liquids including nitric acid solutions (volume 11 800 m<sup>3</sup>) and suspensions (volume 19 000 m<sup>3</sup>). HLW nitric acid solutions are stored in cylindrical tanks with stainless steel covers and bottoms, whereas suspensions are stored in concrete tanks lined with stainless steel. All storage tanks are equipped with level, pressure, temperature, overflow and gas emission monitoring systems.

By 1996, Mayak PA was producing 16 000-20 000 m<sup>3</sup> of liquid intermediate-level waste (ILW) per year with a total activity of less than 30 PBq per annum. On average, the waste has an associated specific activity of 370-750 MBq l<sup>-1</sup> and an average salt content of 12-15 g l<sup>-1</sup>. Currently, liquid ILW is discharged into Lake Karachay and Reservoir 17. The inventory for Reservoir 17 in 1993 was 74 PBq. Lake Karachay has been used as an ILW repository since 1951 and in the period 1951-1993 received 3.55 million m<sup>3</sup> of liquid waste with activity 20 500 PBq. Due to radioactive decay, the content of beta-emitting radionuclides has fallen to a level of 4 400 PBq, in 1996, mainly arising from <sup>90</sup>Sr + <sup>90</sup>Y and <sup>137</sup>Cs. It should also be noted that due to the potential for redistribution of contaminated waters and sediments from the confines of the lake, remediation

measures have been applied in the years after 1967. This has mainly involved the operation of filling in the lake first with gravel and soil and later with hollow concrete blocks. This has resulted in a reduction of the lake area from 0.51 km<sup>2</sup> in 1961 to 0.15 km<sup>2</sup> in 1994. Migration of contaminated groundwater is of obvious concern and monitoring data from Mayak have shown that a volume of  $5 \times 10^6$  m<sup>3</sup> groundwater is contaminated by <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>60</sup>Co and <sup>106</sup>Ru with concentration factors a factor of 20-300 lower than in Lake Karachay.

Liquid low-level waste (LLW) at Mayak PA consist of waters from the facility's sewage system, secondary circuit cooling water from the 2 reactors and cooling water from the high-level waste storage cells. Salts and radionuclides are removed from raw sewage water before discharge with a maximum activity of 7 kBq l<sup>-1</sup> into Lake Kyzyltash. Reservoir 3 in the cascade of storage reservoirs also receives waste from ion exchange processing with maximum activities of 4 MBq l<sup>-1</sup> and a volume of  $0.1 \times 10^6$  m<sup>3</sup> per annum. Over the period 1956-1990, it has been estimated that approximately 6.7 PBq of long-lived radionuclides have entered the reservoirs as low-level wastes most being retained in Reservoir 10.

### 3.1.3 Major releases of radioactivity to the environment from Mayak PA

Three major incidents have led to severe environmental contamination in the vicinity of the production association, namely :

1. Direct discharge of liquid radioactive wastes to the Techa River (1949-1956)
2. The Kyshtym accident in 1957
3. The dessication and transport of contaminated sediments from Lake Karachay in 1967

#### 3.1.3.1 Radionuclide releases to the Techa River

Data relating to direct releases to the Techa River (Academy of Science, 1991; Chukanov *et al.*, 1991; Trapeznikov *et al.*, 1993; Bradley and Jenquin, 1995a; JNREG, 1997). In the following section the data reported in JNREG (1997) are taken as the definitive source of information.

Direct releases of radionuclides were made to the Techa river system between 1949 and 1956. During this period over 100 PBq of radioactive material was discharged into Reservoir 3 which, along with Reservoir 4, acted as a sedimentation pond, before subsequent release into the flowing river (JNREG, 1997; Malyshev *et al.*, 1997). Isotopes of ruthenium (<sup>103</sup>Ru, <sup>106</sup>Ru) and rare earth nuclides formed over 50 % of the total activity and an estimated 12 PBq of <sup>90</sup>Sr and 13 PBq <sup>137</sup>Cs were discharged into the reservoir (JNREG, 1997). According to Russian estimates, approximately 2 TBq of total alpha ( $\Sigma\alpha$ ) were also released to the system at this time. About 98 % of the total activity was attributable to the period from December 1949 to November 1951. It has been estimated that for the period of heaviest discharges (1951), the reduction in the specific activity of the water entering the village of Metlino reached 68 % for <sup>90</sup>Sr and 89 % for <sup>137</sup>Cs, demonstrating the important role of the reservoirs in serving as sedimentation ponds for adsorbed radionuclides. However, the activity which did not precipitate in the reservoirs was still sufficiently high to cause severe contamination along the entire length of the Techa River. About 7 500 people evacuated from 20 settlements along the course of the Techa received radiation doses ranging from 35-1700 mSv (Aklejev & Lyubchansky, 1994). Discharges of <sup>90</sup>Sr and <sup>137</sup>Cs that occurred during the period 1949-1957 have contaminated 240 km<sup>2</sup> of the Techa River flood-plain, with an area of 80 km<sup>2</sup> contaminated at levels above  $3.7 \times 10^{10}$  Bq km<sup>-2</sup> (Bradley and Jenquin, 1995a).

Owing to the high levels of contamination in the upper flood plains of the Techa River, the construction of reservoirs was undertaken in order to contain the activity and act as a storage basin for low level wastes (Bradley and Jenquin, 1995a). Reservoir 10 was completed in 1956 and the discharges of radioactivity to the Techa River, down stream, dropped considerably. The construction of Dam 11, in 1963, further reduced the activity entering the Techa river system. The legacy of this partial containment of the contamination originating from the Mayak PA site are reservoirs which contain high levels of radionuclides such as  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{60}\text{Co}$  and isotopes of plutonium.

### **3.1.3.2 The Kyshtym accident**

A thermal explosion of a tank containing high level liquid waste occurred in September 1957 creating what has become known as the Kyshtym accident. Some 740 PBq was actually released during the accident but an estimated 90 % settled in the immediate vicinity of the explosion site. This meant that 74 PBq of radioactivity was released into the atmosphere and dispersed by the wind to form the East Urals Radioactive trace. Reported levels for the release of radioactivity (Bradley & Jenquin, 1995a; Karavaeva *et al.*, 1994; JNREG, 1997; and Kryshev *et al.*, 1997) appear to have originated from a single official literature source since the reported values agree.

### **3.1.3.3 Wind dispersion from Lake Karachay**

During the period from April 10 to May 15<sup>th</sup> 1967, contaminated sediments from Lake Karachai were dispersed by the wind up to distances of 50-75 km from the Mayak PA site. An estimated 22 TBq was associated with this contamination event (JNREG, 1997). This is the official Russian release estimate as also reported by the Academy of Science (1991). Caesium-137 was the predominant long-lived radionuclide constituting this release and may have accounted for 75 % of the total radioactive inventory (Aarkrog *et al.*, 1997).

### **3.1.4 Present-day contamination levels and correspondence with release estimates**

In the following section, the contamination levels in the vicinity of Mayak PA that had been determined prior to or external from the investigations (sampling) carried out under the framework of the SUCON project are reported. It was felt that these data were best presented in comparison with the official release figures so that discrepancies would be highlighted. It should be clarified that all data extracted from JNREG (1997) relate to earlier Russian measurements as oppose to the measurements made, bilaterally, in 1994 and 1996.

#### **3.1.4.1 Discharges to the Techa River - comparison of release estimates and measurements**

Data relating to contamination in the vicinity of Mayak PA from open literature sources has been assessed. Estimates of present-day inventories are summarised in Table 1.1.

Table 1.1. Inventories in selected environmental compartments - early 1990s.

Location (year)	Total (PBq)	Reference
Asanov Swamp (early 1990s)	0.44 <sup>a</sup>	Bradley & Jenquin (1995a)
Asanov Swamp (1992)	0.25	JNREG (1997)
Reservoir cascade (early 1990s)	7.1	Academy of Science (1991)
Reservoirs 2,3,4, 10 & 11 (early 1990s)	7.4	Bradley & Jenquin (1995a)
Reservoirs 2,3,4, 10 & 11 (1994)	10.5	JNREG (1997)

<sup>a</sup> assuming that <sup>137</sup>Cs and <sup>90</sup>Sr form the main fraction of the total radioactivity

Bradley & Jenquin (1995a) report a total inventory almost double of that reported in JNREG (1997) for the Asanov Swamp whereas the estimate of the total inventory in the reservoir cascade (reservoirs 2,3,4, 10 & 11) are significantly higher in the JNREG (1997) report than in other literature sources.

The Russian data in JNREG (1997) were used to compare the value for the reported release of <sup>137</sup>Cs, virtually all of which occurred between late 1949 and 1951, with the sum of the <sup>137</sup>Cs inventories (decay corrected to 1950) of reservoir 3,4, 10 and 11 and the flood-plain of the Techa River in order to establish whether the values correspond. The decay-corrected inventory calculations are summarised in Table 1.2.

Table 1.2. Present-day <sup>137</sup>Cs inventories decay-corrected to 1950.

Location	Present <sup>137</sup> Cs (PBq)	Decay corrected to 1950	Reference
Reservoir 3 + 4 <sup>a, b</sup>	0.83	2.28	p.129 JNREG (1997)
Reservoir 10 <sup>c</sup>	2.50	6.71	p.105 JNREG (1997)
Reservoir 11 <sup>d</sup>	0.60	1.69	p.105 JNREG (1997)
Techa flood plain + bed <sup>a, e</sup>	0.11	0.29	p.129 JNREG (1997)
<b>Total</b>		<b>11</b>	

<sup>a</sup> assuming <sup>137</sup>Cs makes up the same % of total-β as for Reservoir 10 and 11 (i.e. approximately 44 %, p. 105).

<sup>b</sup> activity level at 1994; <sup>c</sup> activity level at 1993; <sup>d</sup> activity level at 1995; <sup>e</sup> activity level at 1992

It has been assumed that the inventory of <sup>137</sup>Cs residing in the water of the Techa River is negligible relative to the inventories of other environmental compartments. If one assumes the release data to be correct, i.e. 13 PBq released in the early 1950s, 2 PBq of activity are unaccounted for by the inventory estimates presented above. This discrepancy becomes larger still if one also considers the continuous supply of <sup>137</sup>Cs to the system in the period 1958 to present by routine discharges (Section 1.2) which will have had the effect of augmenting the contamination levels in the reservoir cascade. It is tempting to explain the difference by the loss of <sup>137</sup>Cs to the river system below the Techa River but a more likely explanation is provided by the large uncertainties associated with both the discharge data and inventory determinations.

Considering <sup>90</sup>Sr data in JNREG (1997), reported releases of 12 PBq compare to a decay corrected inventory (1950) based on present-day values of 16.2 PBq as shown in Table 1.3.

Table 1.3. Present-day  $^{90}\text{Sr}$  inventories decay-corrected to 1950.

Location	Present $^{90}\text{Sr}$ (PBq)	Decay cor- rected to 1950	Reference
Reservoir 3 + 4 <sup>a,b</sup>	1.02	2.97	p.129 JNREG (1997)
Reservoir 10 <sup>c</sup>	3.90	11.10	p.105 JNREG (1997)
Reservoir 11 <sup>d</sup>	0.58	1.73	p.105 JNREG (1997)
Techa flood plain + bed <sup>a,e</sup>	0.14	0.38	p.129 JNREG (1997)
<b>Total</b>		16.2	

<sup>a</sup> assuming  $^{90}\text{Sr}$  makes up the same % of total- $\beta$  as for Reservoir 10 and 11 (i.e. approximately 54 %, p. 105).

<sup>b</sup> activity level at 1994; <sup>c</sup> activity level at 1993; <sup>d</sup> activity level at 1995; <sup>e</sup> activity level at 1992

The information relating to  $^{90}\text{Sr}$  releases and present-day inventories do not balance although continuous inputs to Reservoir 10 in the period 1958 to present may account for some of the discrepancy. Uncertainties in the modern inventory calculations for Reservoir 10 and 11 are believed to be small since they have reportedly been based on approximately 100 sampling locations in each reservoir. Uncertainties in the present day inventory calculations for Reservoirs 3 & 4 and the Techa River flood-plain are likely to be more significant owing to the assumptions made during calculation. The uncertainties in the release data are believed to be the most likely cause of the discrepancy if one considers that instrumentation for monitoring individual radionuclides, at that time, is likely to have been crude (gross beta measurements were probably taken, assumptions must have been made in relation to exact composition). The calculations here suggest that actual releases may have been at least 4 PBq higher than the official release estimates.

The greatest uncertainties clearly exist for alpha releases to the Techa River. An estimated inventory (1993-1995) of approximately 46 TBq Pu- $\alpha$  is contained in Reservoirs 10 and 11 alone (JNREG, 1997) compared to the official release estimates of 2 TBq  $\Sigma \alpha$ .

To complete the area affected by the direct discharges to the river, some insight is required in relation to the riverine transport of radionuclides to more remote locations. Most evidence in the open literature suggests that the releases of radioactivity to the river system in the early 1950s caused local contamination of the Techa system, with little transport of contamination to sites further downstream. For  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$ , it appears that distinct signals (derived from Mayak) of these radionuclides in sediment profiles from the Ob River (Salekhard on the Ob is some 2500 km from Mayak) may be absent (Panteleyev *et al.*, 1995; Sayles *et al.*, 1997), despite the fact that discharges of these and other radionuclides, to the upper river system, during the early 1950s were very high. However, the scale of the Ob river systems should not be forgotten - inputs of contamination from the River Techa, even if significant in terms of specific activity, are likely to be subject to dramatic dilution from water and suspended sediments carried by the myriad tributaries of the Ob system. It is therefore not surprising that distinct signals cannot be seen some 2 500 km from the source. Other work (Bradley & Jenquin, 1995b) suggests that inputs of  $^{137}\text{Cs}$  to the Barents and Kara Seas from river flow are small compared to global fallout and the influence of European reprocessing plants. According to Mayak scientists, as discussed in Bradley & Jenquin (1995a), 99 % of the contamination, from the period of elevated releases, is deposited upstream from Muslyumovo (78 km downstream from the discharge point). This suggests that no more than 1 % (or

1 PBq) of the total 100 PBq (approximately) discharged reached the lower river system and ultimately the Arctic Ocean. Russian studies cited in Bradley & Jenquin (1995b) estimated that 0.17 PBq (4 700 Ci) of <sup>90</sup>Sr were carried to the Ob River and subsequently to the Kara Sea between 1961 and 1990.

### 3.1.4.2 The Kyshtym accident - correspondence between release figures and reported inventories

The initial radionuclide composition of the accidental release of radioactivity is reported in JNREG (1997). The composition was dominated by short-lived radionuclides (<sup>144</sup>Cs, <sup>95</sup>Zr and <sup>106</sup>Ru) and 2.7 % of long-lived <sup>90</sup>Sr. If one assumes that <sup>90</sup>Sr is the only radionuclide that remains in any significant quantities after approximately 40 years (<sup>137</sup>Cs is reported to have formed only 0.04 % of releases, Pu-isotopes in «trace amounts») then a simple calculation can be made to decay correct the release figure (0.027 x 74 PBq = 2 PBq <sup>90</sup>Sr in 1957) to 1996. This yields a value of 0.77 PBq. According to JNREG (1997), of the activity originally released, an inventory (1996) of 0.75 PBq is associated with the East Urals Radioactive Trace. This value actually reflects the decay-corrected release with corrections made for the activity removed by runoff water, burial or transport into deep soil by deep ploughing. Indeed, after closer inspection an estimated 20 TBq (of which 5.6 TBq was <sup>90</sup>Sr) is reported in JNREG (1997) to have been washed out of contaminated areas into rivers entering the River Sinará basin. A correction to the basic decay-corrected release which accounts for this removal process is consistent with the reported modern day inventory of 0.75 PBq.

A check on the official release data can be derived from the <sup>90</sup>Sr deposition maps (Figure 1.2) and a corresponding table (Table 1.4) that have been produced according to information from Mayak PA. These maps are presented in several reports (Administration of Chelyabinsk Region, 1995; Bradley & Jenquin, 1995a; JNREG, 1997).

Table 1.4. <sup>90</sup>Sr inventory data after initial release from the Kyshtym accident - decay corrected to 1996.

Deposition kBq m <sup>-2</sup>	Area km <sup>2</sup>	<sup>90</sup> Sr inventory		
		min (PBq)	mean (PBq)	Max (PBq)
3.7-74	15 000-20 000*	0.074	0.777	1.48
74-740	600	0.044	0.2442	0.444
740-3700	280	0.2072	0.6216	1.036
3 700-37 000	100	0.37	2.035	3.7
37 000-150 000	17	0.629	1.5895	2.55
	<b>Total (1957)</b>	1.32	5.27	9.21
	<b>Total (1996)</b>	0.51	2.04	3.57

\* 20 000 km<sup>2</sup> used.

It is interesting to note that there are large uncertainties associated with the area contaminated with a deposition in excess of 3.7 kBq m<sup>-2</sup>. In the JNREG (1997) report a range is quoted and a footnote explains that the boundaries for the areas having contamination densities of 3.7 kBq m<sup>-2</sup> (or higher) could not be established reliably. In other studies (Alexakhin *et al.*, 1996; Kryshev *et al.*, 1997) an area of 23 000 km<sup>2</sup> was allocated to this contamination density. For this reason the upper part of the range in JNREG (1997) was used in calculations.

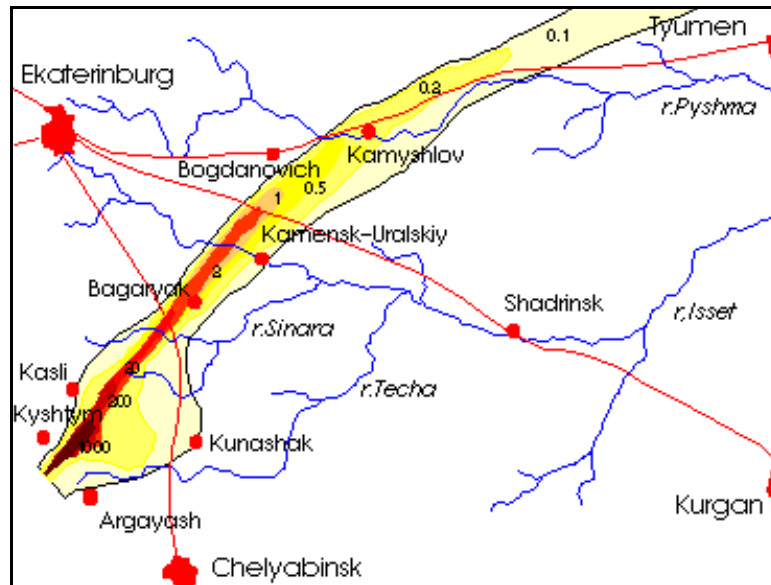


Figure 1.2. The East Urals Radioactive Trace (EURT): Initial contamination densities of  $^{90}\text{Sr}$  ( $\text{Ci km}^2$ ).

The (1957) inventories, based on the original deposition maps, range from 1.32 to 9.21 PBq. If we assume a linear decrease in activity levels between boundaries along both axes (transversal and longitudinal) of the plume (this is a simplification of the Gaussian distribution pattern but acts as a reasonable first approximation), then the middle of the range (referred to as the mean value in Table 1.4) can be taken as the best estimate of the activity levels between boundaries. The (1957) inventory, based on this calculation, yields a value of approximately 5.3 PBq. This is more than twice the official release estimate reported in JNREG (1997) of 2 PBq.

### 3.1.4.3 Lake Karachay incident - correspondence between release figures and reported inventories

A check on the official Russian release figure has been made by considering the map presented in JNREG (1997) showing the level and distribution of  $^{137}\text{Cs}$  after the Karachai incident (Figure 1.3) The contamination map showing,  $^{137}\text{Cs}$  levels, is divided into two parts:

- 1: Area:  $1800 \text{ km}^2$ . Contamination densities in the range:  $11\text{-}210 \text{ kBq/m}^2$
- 2: Area:  $34 \text{ km}^2$ . Contamination densities in the range:  $210\text{-}765 \text{ kBq/m}^2$

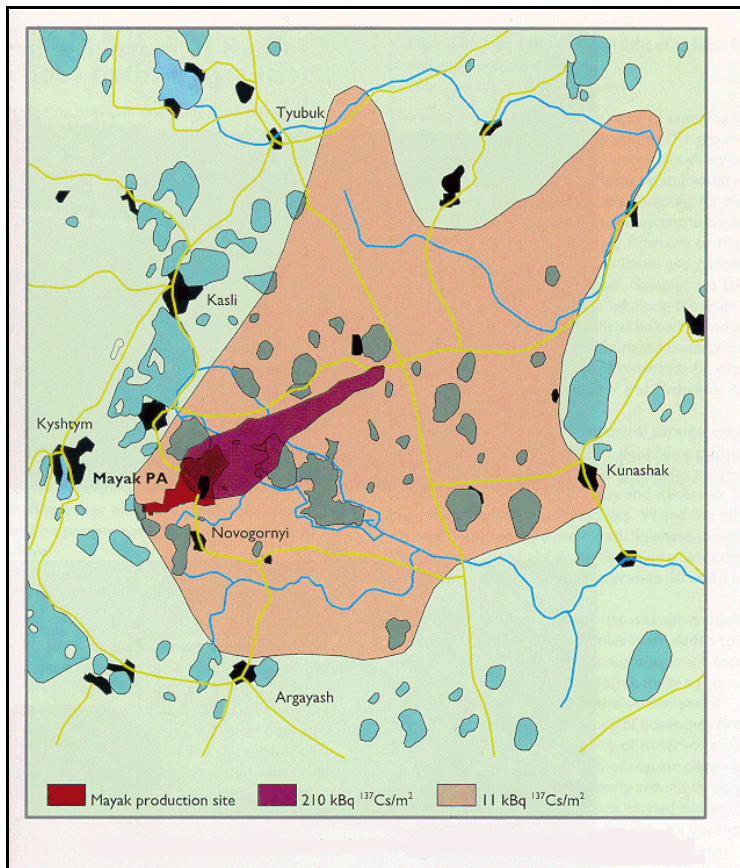


Figure 1.3.  $^{137}\text{Cs}$  contamination from the dispersion of wind-borne contamination from Lake Karachai.

Calculations can be made in a similar fashion to those made for the Kyshtym accident (Table 1.5).

Table 1.5.  $^{137}\text{Cs}$  inventory data after initial release from Lake Karachai.

Deposition kBq m <sup>-2</sup>	Area km <sup>2</sup>	$^{137}\text{Cs}$ inventory		
		Min (PBq)	Mean (PBq)	Max (PBq)
11-210	1800	0.02	0.2	0.38
210-765	34	0.007	0.017	0.026
Total (1967)		0.027	0.22	0.41

The minimum inventory level (27 TBq) is in closest agreement with the reported Russian release (22 TBq). However, this is not the usual way in which this type of data is presented and is not consistent with the text explanation in the JNREG (1997) report. If we, instead, assume a linear fall in activity between the contamination contours in the map then the average inventory (220 TBq) yields a best estimate for the total inventory in 1967. This value is a factor of ten higher than the reported Russian release of  $^{137}\text{Cs}$  from Lake Karachay. Using the mean inventory is also consistent with the maps produced by Administration of Chelyabinsk Region (1995). It should be noted that the contribution of  $^{137}\text{Cs}$  from the 1957 Kyshtym accident can not explain the high inventories calculated, i.e. using the  $^{90}\text{Sr}$  contamination map given in the Administration of Chelyabinsk Region (1995) report, and assuming a  $^{90}\text{Sr}/^{137}\text{Cs}$  ratio of 67, the contribution of  $^{137}\text{Cs}$  from the Kyshtym accident is in the range 0.01-0.5 kBq/m<sup>2</sup>



in area 1 (as defined above), levels which are insignificant when considering the unit inventories ( $> 11 \text{ kBq m}^{-2}$ ) in the area. Decay-correcting the 220 TBq  $^{137}\text{Cs}$  calculated above to 1996 gives an inventory of approximately 110 TBq.

## 3.2 Present Levels of Contamination and Source Identification.

### 3.2.1 Present levels of contamination and source identification in the Mayak PA near-zone

Data from the joint Russian-Norwegian fieldwork of 1994 and 1996 form an important input to the SUCON project and have been presented in several publications and conference proceedings (Christensen *et al.*, 1995; Tronstad *et al.*, 1995; Salbu *et al.*, 1997; JNREG, 1997). The following section provides a summary of some of the main results and conclusions, relating to levels of radionuclide contamination in soil, sediment waters and biota, and source identification from these studies. It should be noted that at the time of writing of this report, all data from the 1994 field campaign have been compiled and published in one report (JNREG, 1997), whereas data from 1996 have only been selectively reported.

#### 3.2.1.1 Contamination levels in the Mayak PA near-zone

Sampling locations for the 1994 joint Russian-Norwegian fieldwork are shown in Figure 2.1.

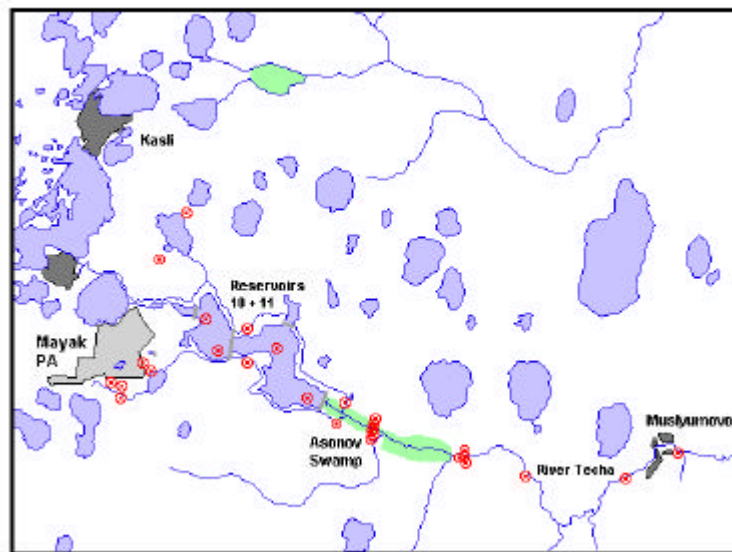


Figure 2.1. Location of sampling sites in the joint Norwegian-Russian study of 1994.

##### 3.2.1.1.1 Contamination levels in soils

A summary of contamination densities for soils collected in the vicinity of Mayak PA is presented in Table 2.1. All measured activity levels for  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in surface soils showed good agreement with Russian estimates of contamination densities in the Mayak area. Highest levels of  $^{90}\text{Sr}$  contamination were observed for those sites in the EURT ( $14\text{-}34 \text{ MBq m}^{-2} \text{ }^{90}\text{Sr}$ ) and highest levels of

$^{137}\text{Cs}$  contamination were observed in the Asanov Swamp at a distance of 7 km from Dam 11 (42 MBq m<sup>-2</sup>). Even at the inhabited village of Muslyumovo, some 47 km from the industrial reservoirs, contamination densities of 10 MBq m<sup>-2</sup>  $^{137}\text{Cs}$  were determined.

Table 2.1. Contamination densities (kBq m<sup>-2</sup>) for soils (1994) in the vicinity of Mayak PA (adapted from JNREG, 1997).

Sampling location	Contamination density (kBq m <sup>-2</sup> )				
	<sup>60</sup> Co	<sup>90</sup> Sr	<sup>137</sup> Cs	Σ Pu	<sup>241</sup> Am
Repository site	<2.8 - 190	3 400 - 11 000	1 200 - 3 800	71 - 120	45 - 100
Health Protection zone	<0.05 - 0.075	97 - 180	210 - 380	2.3 - 2.4	1.0 - 4.3
Asanov swamp	b.d.l.	130 - 10 000	400 - 42 000	1.1 - 85	n.a.
Techa river	b.d.l.	62 - 350	470 - 2 800	0.4 - 7.0	n.a.
Muslyumovo	<0.01 - <0.03	1 000	4 700 - 10 000	13 - 29	<0.1 - <1.3
EURT	2.3 - 3.7	14 000 - 34 000	530 - 1 400	12 - 17	5.9 - <11

b.d.l. = below detection limits; n.a. = not analysed; Σ Pu = sum of alpha-emitting Pu isotopes

### 3.2.1.1.2 Contamination levels in groundwater, reservoir water and river water

Filtered water samples were collected from 2 boreholes in the vicinity of Lake Karachay in 1994. At a borehole 3.6 km SE of the lake, levels of 4 200 Bq l<sup>-1</sup> <sup>60</sup>Co and 8 800 Bq l<sup>-1</sup> <sup>90</sup>Sr were determined at depths below 50 m. Caesium-137 activity levels were below 2 Bq l<sup>-1</sup> at this site. At a borehole only marginally further from Lake Karachay (4 km South) levels of activity were considerably lower with maximum levels of 270 Bq l<sup>-1</sup> <sup>60</sup>Co and 68 Bq l<sup>-1</sup> <sup>90</sup>Sr. Data from the regular monitoring of this second borehole show a steady increase in <sup>90</sup>Sr levels in the period 1989-1994.

Surface waters were also sampled from the Mishelyak River, Reservoirs 10 and 11, the by-pass channels and Techa River waters in 1994. Highest levels of activity were measured in Reservoir 10 (8.4 - 14 kBq l<sup>-1</sup> <sup>90</sup>Sr, 100 Bq l<sup>-1</sup> <sup>137</sup>Cs, 1.5-1.8 Bq l<sup>-1</sup> <sup>60</sup>Co) with levels falling significantly in Reservoir 11 (1.9 - 2.4 kBq l<sup>-1</sup> <sup>90</sup>Sr, 1.1-1.5 Bq l<sup>-1</sup> <sup>137</sup>Cs, <0.1 Bq l<sup>-1</sup> <sup>60</sup>Co). Levels of activity in the Techa River, which primarily receives radionuclide contamination from the reservoir cascade *via* filtration of water into the by-pass channels and through the wall of Dam 11, were fairly constant between the Asanov Swamp, 15 km from Dam 11 and Muslyumovo, some 47 km downstream of Dam 11. Levels of 7-10 Bq l<sup>-1</sup> <sup>90</sup>Sr and 0.6-0.8 Bq l<sup>-1</sup> <sup>137</sup>Cs were reported. Activity levels from reservoir waters analysed in 1996 were consistent with the levels reported in 1994.

### 3.2.1.1.3 Contamination levels in reservoir and river sediments

A summary of contamination densities for sediments collected in the vicinity of Mayak PA is presented in Table 2.2. The bed of both Reservoirs 10 and 11 are comprised of both sediments from the old Techa River bed/floodplains and soils flooded after the construction of the reservoirs. It is, therefore, not surprising that a large range in contamination densities exists for these sediments. Caesium-137 and <sup>90</sup>Sr were found to be the most important radionuclides in terms of their contribution to the total inventory in these reservoirs. Contamination densities of up to 21 MBq m<sup>-2</sup> <sup>137</sup>Cs were measured in Reservoir 10. High contamination densities were also recorded in the Asanov Swamp (7 km downstream of Dam 11) with reported values of 44 MBq m<sup>-2</sup> <sup>137</sup>Cs and 1.2 MBq m<sup>-2</sup> <sup>90</sup>Sr. Contamination densities decreased by a factor of 40 for <sup>137</sup>Cs and a factor of 10 for <sup>90</sup>Sr between this sampling location and Muslyumovo. The 11 cores

collected in the reservoirs in 1996 provide a further indication of the heterogeneity of the sediments and a verification Russian inventory estimates.

Table 2.2. Contamination densities ( $\text{kBq m}^{-2}$ ) for sediments (1994) in the vicinity of Mayak PA (adapted from JNREG, 1997).

Sampling location	Contamination density ( $\text{kBq m}^{-2}$ )				
	$^{60}\text{Co}$	$^{90}\text{Sr}$	$^{137}\text{Cs}$	S Pu	$^{241}\text{Am}$
Reservoir 10	210-530	4 800 - 9 500	6 100 - 21 000	470-720	270-860
Reservoir 11	6-90	8 100 - 13 000	170 - 14 000	0.2-63	25
Asanov Swamp	1.8	1200	44 000	74	n.a.
R. Techa (15 km from Dam 11)	b.d.l.	n.a.	880	0.5	b.d.l.
R. Techa (25 km from Dam 11)	b.d.l.	n.a.	180	0.2	b.d.l.
R. Techa (42 km from Dam 11)	b.d.l.	50	2000	1.5	b.d.l.
R. Techa (47 km from Dam 11)	b.d.l.	110	1100	0.55	b.d.l.

b.d.l. = below detection limits; n.a. = not analysed;  $\Sigma$  Pu = sum of alpha-emitting Pu isotopes

### 3.2.1.1.4 Contamination levels in Biota

Samples of vegetation and fish were analysed following the field campaign of 1994 and 1996. Transfer factors (activity per dry weight vegetation/ Specific activity per dry weight soil) were typically 0.3 for  $^{90}\text{Sr}$  and 0.04 for  $^{137}\text{Cs}$ .

Fish samples captured in Reservoirs 10, 11 and the Techa River were also analysed after the field-work of 1994 and have been the subject of a recent article (Strand *et al.*, submitted). Activity concentrations up to  $140 \text{ kBq kg}^{-1}$  (f.w.)  $^{137}\text{Cs}$  in pike (*Esox lucius*) were recorded in Reservoir 10 although levels in the Techa River, at Muslyumovo, were much lower, i.e.  $400 \text{ Bq kg}^{-1}$  (f.w.)  $^{137}\text{Cs}$ . Caesium-137 Concentration Factors (CFs - Specific activity per wet weight/ Specific activity per litre) as high as  $1\,400 \text{ l kg}^{-1}$  were calculated for pike from Reservoir 10. A «trophic level» effect was evident in Reservoir 11 (pike CF=1000, roach, *Rutilus rutilus* CF = 240). The fact that  $^{137}\text{Cs}$  CFs fall towards the lower end or below the expected range, may be reflect the presence of highly-mineralised waters.

### 3.2.1.2 Source identification in the Mayak PA near-zone

The contamination of the area surrounding Mayak PA can be attributed to a number of source terms as discussed in some detail in Chapter 1. To briefly recapitulate these include the direct discharges to the Techa River (1949-56), the discharges to the reservoirs (1956-present), discharges to Lake Karachay (1951-present), Kyshtym accident (1957), dessication of lake Karachai and wind-blown dispersion of contaminated silt (1967) and airborne releases from plant stack. These source terms vary with respect to activity levels, radionuclide composition (and hence ratios) and physico-chemical forms. Furthermore, ratios may provide information on industrial activities. An example of the use of ratios is the consideration of  $^{240}\text{Pu}/^{239}\text{Pu}$  data to consider sources. Weapons plutonium can be easily distinguished from that arising from global fallout and civil uses by this technique. Weapons-grade plutonium requires a high content of the fissionable  $^{239}\text{Pu}$ -isotope and is therefore characterised by relatively low  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios, usually less than 0.05. In contrast, both global weapons fallout and spent fuel from civil reactors have higher  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios due to neutron activation of  $^{239}\text{Pu}$ . Activity ratios for the main sources and for samples analysed after the 1994 field campaign are presented in Table 2.3.

Table 2.3. Summary of activity ratios for sources (official release figures) and samples collected at Mayak in 1994 (Mean  $\pm$  SEM).

Source/site	$^{90}\text{Sr}/^{137}\text{Cs}$	$\Sigma\alpha/^{137}\text{Cs}$ <sup>a</sup> $\Sigma\text{Pu-}\alpha/^{137}\text{Cs}$	$^{238}\text{Pu}/^{239,240}\text{Pu}$ (Activity ratio)
<b>Discharge to Techa, (1949-1956)</b>	<b>0.92</b>	<b>0.00015</b>	
Reservoir 10	0.32 $\pm$ 0.01, 1.5 <sup>b</sup>	0.042 $\pm$ 0.008	1.4 $\pm$ 0.2
Reservoir 11	1.2 $\pm$ 0.1, 31 <sup>b</sup>	0.0039 $\pm$ 0.004	0.75 $\pm$ 0.21
Techa River sediments (3-47 km)	0.06 $\pm$ 0.02	0.0010 $\pm$ 0.0003	0.018 $\pm$ 0.003
Techa River soils (3-47 km)	0.20 $\pm$ 0.02	0.0024 $\pm$ 0.0004	0.0051 $\pm$ 0.0003
<b>Kyshtym accident (1957)</b>	<b>67</b>		
EURT soils, 15-20 km	26	0.025	0.011 $\pm$ 0.002
<b>Discharges to Lake Karachay</b>	<b>0.21</b>	<b>0.0004<sup>c</sup></b>	
Groundwater	2 0.3		0.77
<b>Wind dispersion from Karachay (1967)</b>	<b>0.34</b>	<b>0.0004<sup>c</sup></b>	
Waste burial sites, soil	3.3 $\pm$ 0.3	0.14 $\pm$ 0.04, 0.02 <sup>b</sup>	0.07 $\pm$ 0.01
Health Protection Zone, soil	0.46 $\pm$ 0.04	0.017 $\pm$ 0.005	0.44
R, Mishelyak (0-3 cm) sediment	22	0.011	
(3-38 cm) sediment	0.45	0.013	

<sup>a</sup> = for sources, no data were available relating to  $\Sigma\text{Pu-}\alpha$  therefore values related to total alpha measurements

<sup>b</sup> = variation between sampling sites was significant

<sup>c</sup> = Ratio for Lake Karachay sediments in 1970.

(a) EURT : The  $^{90}\text{Sr}/^{137}\text{Cs}$  ratios for soil samples collected from the head of the EURT in 1994 (26) were significantly lower than the  $^{90}\text{Sr}/^{137}\text{Cs}$  ratios derived from the official release figures (67). Adjustment for the enhancement of  $^{137}\text{Cs}$  in these samples due to the influence of deposition from Lake Karachay cannot account for the discrepancy (Brown and Amundsen, 1998). An assessment of the available evidence suggests that either releases of  $^{137}\text{Cs}$  were higher or  $^{90}\text{Sr}$  releases lower than the official release figures or that there were other unrecorded contamination events in the area. Other possible explanations relate to the fractionation of radionuclides in the environment. It is known that  $^{90}\text{Sr}$  is more mobile than  $^{137}\text{Cs}$  in soils which may have resulted in the leaching of a larger proportion of this radioisotope to lower layers below the depth of sampling.

(b) Wind dispersion of contaminated sediment from Lake Karachay : In soils collected to the South of the lake,  $^{90}\text{Sr}/^{137}\text{Cs}$  ratios were comparable to the ratios officially reported for Lake Karachay fallout. However,  $\text{Pu-}\alpha/^{137}\text{Cs}$  ratios were significantly higher than those reported for Lake Karachay sediments in 1970 suggesting an additional source of plutonium to the sampling area.

(c) The Techa River and Asanov Swamp : The  $^{90}\text{Sr}/^{137}\text{Cs}$  ratios in river sediments and soils are significantly lower than those observed in reservoir sediments although the main source of contamination, i.e. direct discharges of ILW to the River Techa (1949-56), are believed to be the same. There is evidence, from inventory estimates and river concentration data, that a significant fraction of the  $^{90}\text{Sr}$  deposited during the high activity release period may have been lost (at least for the period 1962-1992) from the Asanov Swamp whereas  $^{137}\text{Cs}$  has remained in a more immobile form. The low  $^{90}\text{Sr}/^{137}\text{Cs}$  ratios in the lower Techa support this theory. The  $^{238}\text{Pu}/^{239,240}\text{Pu}$  ratios in river soils and sediments are significantly lower than those observed in the reservoirs which supports the

contention that the reservoirs received an additional source of  $^{238}\text{Pu}$  after the damming of the Techa River.

(d) Reservoirs 10 and 11 : The  $\text{Pu-}\alpha/^{137}\text{Cs}$  ratios in the sediments of Reservoir 10 were a factor of 10 higher than in Reservoir 11, and both ratios are significantly higher than the ratio reported for the discharges in 1949-1956. Radiocesium and plutonium exhibit quite different biogeochemical behaviour which may explain differences in the  $\text{Pu-}\alpha/^{137}\text{Cs}$  ratios of the 2 reservoirs. The differences in the  $\text{Pu-}\alpha/^{137}\text{Cs}$  ratios between the official discharge figures and the reservoirs can be explained by an additional source of Pu-isotopes after the construction of Reservoirs 10 and 11. Indeed, the presence of the short-lived radionuclides  $^{60}\text{Co}$  and  $^{134}\text{Cs}$  provides evidence for fairly recent waste inputs to the reservoirs. The primary reason for the differences between source and sample ratios, however, is believed to be an under-estimation of Pu in the official release figures as discussed in Chapter 1. Ratios based on the isotopes of Pu provide a far superior marker of sources because isotopes are virtually geochemically identical and should therefore not suffer from the problems of fractionation in the environment. The differences in the  $^{238}\text{Pu}/^{239,240}\text{Pu}$  ratios between reservoirs provides some evidence to suggest divergent histories in terms of waste disposal inputs.

In addition to the determination of  $^{240}\text{Pu}/^{239}\text{Pu}$  made by ICP-MS, determinations by accelerator mass spectrometry (AMS) have also provided valuable additional information in the process of source identification and has been the focus of a recent article produced by SUCON participants (Oughton *et al.*, submitted). The samples from the bilateral campaigns of 1994 and 1996 have been used for this work. Relatively high ratios were observed in Reservoirs 10 and 11 in agreement with a halt in the production of weapons' grade Pu and increased focus on civil reprocessing at the plant. Data from this study also suggested that most of the plutonium in the Upper Techa River originates from early waste discharges. However, enhanced atom ratios in surface sediments at Muslyumovo and sites downstream were considered to possibly indicate the influence of additional, later sources.

### 3.2.2 The zone outside the Mayak controlled area

#### 3.2.2.1 Environmental radioactivity in soil samples outside the nearzone of Mayak

In 1997 a paper on "Radioactive inventories from the Kyshtym and Karachay accidents: Estimates based on soil samples collected in the South Urals (1990-1995)" was published in "The Science of the Total Environment".

At 25 locations located between 7 and 126 km from "Mayak" soil samples were collected to a depth of 30 cm divided in 5 cm sections and analysed for  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{239,240}\text{Pu}$ ,  $^{238}\text{Pu}$ ,  $^{241}\text{Am}$  and other radionuclides if present. Furthermore grass and litter samples were analysed from most of the locations for the same radionuclides as the soil. From the vertical distribution of the various radionuclides it can be concluded that for all locations except one essentially all activity was found in the upper 30 cm. In case of  $^{90}\text{Sr}$ , the 25-30 cm layer contained on the average 1.5% of the total inventory of  $^{90}\text{Sr}$  in the 0-30 cm layer, vegetation included. In the case of  $^{137}\text{Cs}$  and transuranics (Pu and Am) even less was found in the deeper parts of the soil column.

One location was special because at this site the contaminated top soil layers were displaced to a greater depth after the Kyshtym accident in 1957. Hence the

highest  $^{90}\text{Sr}$  concentrations, representing the Kyshtym debris are found in a depth of 50-100 cm at this location.

In order to estimate the contributions of the various radionuclides from the Kyshtym and Karachay accidents a number of assumptions were made. Firstly the background of global fallout from nuclear weapons testing should be established. Two locations (Rassoka) and (Miassovo) were both considered to represent global fallout background with regard to  $^{90}\text{Sr}$ . None of these locations received fallout from the Kyshtym accident in 1957 or from the Karachay dispersion in 1967. They may however, have been slightly contaminated with radiocaesium from the Chernobyl accident in 1986. Neither can some contamination with radiocaesium from MAYAK at Miassovo and by the neighbouring Belyarsk nuclear power plant in the case of Rassoka be excluded. The enhanced  $^{137}\text{Cs}/^{90}\text{Sr}$  ratios (2.5 at Miassovo and 3.2 at Rassoka) do in fact suggest some non global fallout radiocaesium at both locations, as the  $^{137}\text{Cs}/^{90}\text{Sr}$  ratio in global fallout is 1.6.

In the calculations below the deposition found at Rassoka i.e. 1.6 kBq  $^{90}\text{Sr}$  has been used as the global fallout background in the South Urals. The global fallout background of  $^{137}\text{Cs}$  is calculated as 1.6 times the  $^{90}\text{Sr}$  deposition and thus becomes 2.56 kBq  $\text{m}^{-2}$ . The ratio  $^{239,240}\text{Pu}/^{90}\text{Sr}$  in integrated fallout from nuclear weapons testing (corrected for decay of  $^{90}\text{Sr}$ ) was 0.0367 in 1991 and the fallout background is thus calculated to 0.06 kBq  $^{239,240}\text{Pu}$   $\text{m}^{-2}$ . The fallout background of  $^{241}\text{Am}$  becomes 0.025 kBq  $^{241}\text{Am}$   $\text{m}^{-2}$ .

The  $^{90}\text{Sr}/^{137}\text{Cs}$  ratio in Kyshtym debris was 71 and that of the deposition from the Karachay dispersion showed a  $^{90}\text{Sr}/^{137}\text{Cs}$  ratio of 0.3. These assumptions were based on information given by Nikipelov *et al.* (1990)

A set of 4 equations were set up for each location and solved in order to calculate the contributions of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  from the Kyshtym accident and the Karachay dispersion respectively:

$$x + y = a; v + p = b; -71x + v = 0; -0.3y + p = 0$$

$x$ : kBq  $^{137}\text{Cs}$   $\text{m}^{-2}$  from Kyshtym  
 $y$ : kBq  $^{137}\text{Cs}$   $\text{m}^{-2}$  from Karachay  
 $v$ : kBq  $^{90}\text{Sr}$   $\text{m}^{-2}$  from Kyshtym  
 $p$ : kBq  $^{90}\text{Sr}$   $\text{m}^{-2}$  from Karachay

$a$ : is the total measured  $^{137}\text{Cs}$  deposition at the location in kBq  $\text{m}^{-2}$  minus the contribution from global fallout and minus the contribution from other sources e.g. Chernobyl determined from the  $^{134}\text{Cs}$  content.

$b$ : is the total measured  $^{90}\text{Sr}$  deposition at the location in kBq  $\text{m}^{-2}$  minus the contribution from global fallout.

In order to calculate the unknown ratios: ( $^{239,240}\text{Pu}/^{137}\text{Cs}$ ) in Karachay debris and ( $^{239,240}\text{Pu}/^{90}\text{Sr}$ ) in Kyshtym debris, seven locations were selected for the first ratio and six for the second. The selection was based on the above calculations of the contributions of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  from the two accidents and expressions similar to those derived below for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  were calculated for Pu.

In order to calculate the radionuclide inventories from the Kyshtym and Karachay accidents it was assumed that the  $^{90}\text{Sr}$  deposition in kBq  $\text{m}^{-2}$  from the Kyshtym accident followed an exponential decrease with distance from MAYAK and that the activity is confined within a  $15^\circ$  sector out to a distance of 300 km, which approximately describes the contamination pattern as e.g. shown by Romanov *et al* (1990).

Also in the case of Karachay debris an exponential decrease of kBq  $^{137}\text{Cs}$   $\text{m}^{-2}$  with distance was assumed that the debris was confined within a  $60^\circ$  sector out

to a distance of 150 km from MAYAK. This model was derived from the contamination pattern shown by Izrael *et al* (1993) and Tsurov and Anisimova (1993).

For the  $^{90}\text{Sr}$  deposition from the Kyshtym accident the expression becomes:  
 $y = e^{(a+bx)}$ , where

$$a = 8.08 \pm 0.35, \quad b = -0.048 \pm 0.015$$

$y$  is  $\text{kBq } ^{90}\text{Sr m}^{-2}$  and  $x$  = distance from MAYAK in km.

The  $^{90}\text{Sr}$  inventory (I) was calculated for the distance interval 30 to 300 km

$$I = 15/360 \cdot 2\pi \cdot 3240 \cdot 10^{-6} \int_{30}^{300} x \cdot e^{-0.048x} dx = 0.2 \text{ PBq } ^{90}\text{Sr}.$$

The uncertainty of the integral is about a factor of 2.5 i.e. the 1 SD range is 0.1 - 0.5 PBq.

The theoretical deposition from the Kyshtym accident should be about 0.75 PBq  $^{90}\text{Sr}$  in 1996 and of this at least half was deposited within 30 km from MAYAK (Romanov *et al.* 1990). Hence the estimate of 0.2 PBq between 30 and 300 km is not incompatible with the earlier data.

For the  $^{137}\text{Cs}$  deposition from the Karachay incident the expression becomes:  
 $y = e^{(a+bx)}$  where

$$a = 3.68 \pm 0.144, \quad b = -0.021 \pm 0.0048$$

$y$  =  $\text{kBq } ^{137}\text{Cs m}^{-2}$  and  $x$  = distance from MAYAK in km.

The  $^{137}\text{Cs}$  inventory (I) was calculated for the distance interval 7 to 150 km:

$$I = 60/360 \cdot 2\pi \cdot 38 \cdot 10^{-3} \int_7^{150} x e^{-0.021x} dx = 72 \text{ TBq } ^{137}\text{Cs}$$

The uncertainty of the integral is about a factor of 1.5 i.e. the 1 SD range is 50 - 110 TBq.

The total released activity from the Karachay accident in 1967 was 600 Ci (Academy of Science, 1991). If 75% of this was  $^{137}\text{Cs}$ , i.e. 450 Ci this would now, 30 years later, have decayed to 225 Ci or 8.3 TBq. In WP1 the estimated inventory is 7 TBq. But from fig. 4.9 the Joint-Norwegian-Russian Expert Groups report (see ref. to WP1) it can be calculated that the  $^{137}\text{Cs}$  contamination from the Lake Karachay accident has been at least 40 TBq. Hence there seems to be an inconsistency between the Russian release data and actual measurements. The present estimate is nearly an order of magnitude higher. A comparison between the present data and Russian aerial survey measurements in 1991 (Izrael *et al.* and Anisimova 1993) show reasonable agreement southward of MAYAK, but the levels NE of MAYAK were in general twice as high as the Russian 1991 aerial data. It thus seems that the  $^{137}\text{Cs}$  contamination from the Karachay incident may either have been somewhat higher than earlier assumed or there may have been other contamination events in the South Urals than those reported so far. Anyway the soil data indicate higher  $^{137}\text{Cs}$  levels than the Russian aerial data.

The mean  $^{239,240}\text{Pu}/^{90}\text{Sr}$  ratio in Kyshtym debris was estimated to  $0.0018 \pm 0.0007$ . If the estimated  $^{90}\text{Sr}$  Kyshtym inventory is multiplied with this ratio the Pu inventory becomes 0.4 TBq  $^{239,240}\text{Pu}$ . If the total inventory of  $^{90}\text{Sr}$  from Kyshtym is 0.75 PBq the total  $^{239,240}\text{Pu}$  inventory from Kyshtym becomes  $1.8 \pm 0.5$  TBq.

As a mean estimate for the Karachay derived plutonium  $0.7 \pm 0.2$  TBq  $^{239,240}\text{Pu}$  was found. It thus seems that the inventories of  $^{239,240}\text{Pu}$  from the Kyshtym and Karachay accidents both are in the order of 1 TBq.

At the three locations, which contained the highest contamination levels with Kyshtym debris the mean  $^{238}\text{Pu}/^{239,240}\text{Pu}$  ratio in the total deposit was  $0.0115 \pm 0.024$  ( $\pm 1$  SD;  $N = 3$ ) and the  $^{241}\text{Am}/^{239,240}\text{Pu}$  ratio was  $0.117 \pm 0.027$  ( $\pm 1$  SD;  $N = 3$ ). These ratios are about three times lower than those found in global fall-out from nuclear weapons testing. They are also low compared with those found in fuel elements in nuclear power reactors. The low ratios suggest “low burn up” in the nuclear fuel, which is characteristic for nuclear weapons plutonium.

The three mostly contaminated locations with Karachay debris, showed  $^{238}\text{Pu}/^{239,240}\text{Pu} = 0.088 \pm 0.069$  and  $^{241}\text{Am}/^{239,240}\text{Pu} = 0.169 \pm 0.026$ . These ratios are higher than from those in the Kyshtym debris and suggest thus a higher burn-up for the Pu deposited in Lake Karachay.

### 3.2.3 SUCON intercalibration exercise

An intercalibration exercise was undertaken by the laboratories involved in handling SUCON samples to ensure consistency of measurements. A bulk sample of river bed sediment, of predominantly clay-type composition, was prepared by the Institute of Energy Technology (IFE), Norway. The radioactivity in the sediment originates mainly from a uranium reprocessing plant shut down in 1968. The concentrations of the different radionuclides in the sediment vary considerably depending on the depth in the sediment with the highest concentrations currently found about 80 cm below the sediment surface. The preparation process involved dashing the sediment at a temperature of  $450\text{ }^{\circ}\text{C}$ , followed by sieving and mixing of the dried sample. An initial homogeneity of around 6% (relative standard deviation) was believed to have been achieved, based on a series of one hour caesium-137 to americium-241 were used to estimate approximate concentrations of strontium-90 and plutonium-239 in the samples.

Six 25 g aliquots were taken from the bulk sample and provided to the other laboratories by IFE in September 1997 for intercomparison measurements to be carried out.

Table 2.4 provides a summary of the results received from the participating laboratories. All results have been decay corrected to a reference date of 1<sup>st</sup> October 1997.

Table 2.4. Intercomparison results.

Institute	Concentration / Bq kg <sup>-1</sup> (dry mass)		
	Sr-90	Cs-137	Pu-239,240
RISØ, Denmark	20 (3.1)	170 (6.1)	803 (197)
AUN, Norway	19.8 (1.3)	190 (19)	947 (164)
IFE, Norway	37 (15)	180 (7)	951 (135)
IPAE, Russia		289 ± 15.2	
IBSS, Ukraine			612 ± 81.4
Range	17.4 - 59	163 - 289	580 - 1100

Numbers in parentheses indicate one actual standard deviation indicates uncertainty on single measurement reported.

The results generally show a good level of agreement between institutes. The range of results obtained cover the approximate radionuclide content originally estimated by IFE (20 Bq kg<sup>-1</sup> strontium-90, 200 Bq kg<sup>-1</sup> caesium-37, 1000 Bq kg<sup>-1</sup> plutonium-239, 240).



Some specific difficulties have been identified as a result of exercise.

- All institutes agreed that the sub-samples were heterogeneous with respect to plutonium. This is apparent from the large range of results obtained from plutonium-239, 240 analysis and exceeds the initial estimate of homogeneity is radionuclide dependent.  
If the plutonium-239, 240 result from IBSS is compared with the mean results reported by Risø, AUN and IFE, it appears to be substantially lower. However, if the range of results obtained is considered, the IBSS value falls within this range.
- The result of caesium-137 analysis by IPAE exceeds the range of values obtained by the other institutes (163 - 206 Bq kg<sup>-1</sup>). This may be attributed to the relatively small amount of sample used by IPAE in the caesium-137 analysis here compared with other intercomparison exercises carried out between Risø and IPAE which have shown much closer agreement.
- The mean result of the Sr-90 analysis by IFE is approximately two times greater than those obtained by Risø and AUN. The range of results obtained by IFE appears large, although uncertainties on individual measurements were relatively low. These uncertainties are the total error for each sample and represent the errors from the analytical process only. Two more parallels were measured by IFE once the initial results of the intercomparison were known. The results were 21 and 22 Bq kg<sup>-1</sup>, in good agreement with other results. There is no indication from the laboratory records that the analytical work on the subsamples that gave the relatively high results had failed but it is possible that these subsamples do not represent the total sample as well as the other subsamples.

### 3.3 Radiological Dose Assessment

The aim of this part of the project was to carry out a comprehensive dose assessment for population groups in the Southern Urals. As previously discussed the nature of the contamination and the levels of exposure vary throughout the region. Attention was, therefore, focused on particular villages in order to obtain a picture of radiation exposures in the region. A detailed assessment of radiation doses was carried out for the village of Brodokalmak, situated on the Techa river. In addition, doses for the village of Muslyumovo, which is also situated on the Techa river but where levels of contamination are higher than at Brodokalmak, were considered. A dose assessment was also carried out for the village of Bagaryak situated in the East Urals Trace (the area affected by the Kyshtym accidental release). Current and future doses were determined for individuals who are most exposed and for those exposed at more average levels. The various dose assessments are outlined in the following sections and are discussed in more detail by Cabianca et al [Cabianca 1999(a) and (b)].

#### 3.3.1 The Techa River Population

##### 3.3.1.1 Dose assessment methodology

The main assessment was carried out for the village of Brodokalmak situated on both sides of the Techa river about 110 km from the radionuclide release point. The village of Muslyumovo was also considered which is located closer to the MAYAK plant than Brodokalmak, about 78 km from the release point. Several restrictions regarding the use of the Techa River and surrounding areas were

introduced and are still in place at Brodokalmak and Muslyumovo. The restrictions include bans on drinking river water, fishing and bathing in the river; access to the river banks and use of pasture along the river banks are also prohibited. At Brodokalmak, farmers using these areas were allocated pastures in uncontaminated areas away from the river. A bridge across the Techa river connects the village to these areas. It seems that there is not strict control over the application of these measures at present and that the population still uses the Techa river and its banks to some extent to graze cattle and for fishing.

For Brodokalmak a preliminary, deterministic assessment of current doses was carried out [Cabianca 1999(a) and (b)] using 'best estimate' values for the various parameters, this was then followed by a probabilistic assessment taking into account the uncertainties associated with the parameter values [Cabianca 1999(b)]. In both cases doses were estimated for three age groups: infants aged 1 year, 10 year old children and adults. The doses estimated are the sum of the committed effective doses due to the ingestion of radionuclides in a year and the external effective radiation dose received in a year as defined by the International Commission on Radiological Protection [ICRP 1990] for comparison with the annual dose limits. Committed effective doses due to internal irradiation are integrated to age 70 years for infants and children and for 50 years for adults. The doses are, therefore, not the doses actually received in a year but those that the population are committed to. Two scenarios were considered in the calculations. In the first scenario it was assumed that the restricted access to the river banks for both people and cattle was observed by the population. For the second scenario, doses were calculated assuming that the restrictions were lifted and people had free access to all areas in the village, including using the river for fishing and obtaining drinking water. Doses due to ingestion of vegetables, chicken, eggs, bread and cereals remain unaffected by the lifting of the restrictions, because crops are not grown on the river flood plains.

In 1996 a survey of the habits of the population of Brodokalmak (pop. 3600) was carried out by the Institute of Plant and Animal Ecology (IPAE) of Yekaterinburg. The data collected included the daily intake rates of terrestrial and aquatic foods and the number of days per week when these foods were consumed. Annual ingestion rates were calculated from these data with foods grouped together in broad categories to facilitate the calculation of doses. The arithmetic mean value of the intakes of the highest 10% of consumers was adopted as the ingestion rate of the most exposed group. The arithmetic mean value of the intakes for the remaining consumers was used as the average ingestion rate for the Brodokalmak population. The ingestion rates adopted in the assessment are reported in Table 3.1. No local data were available to estimate annual occupancy rates for the river bank or the bridge crossing the Techa river. Occupancy rates were therefore based on generalised UK data [Robinson 1996] taking into account information on the activities of the population of Brodokalmak and the weather conditions for the region. Occupancy rates adopted in the calculations are given in Table 3.2.

Table 3.1. Annual ingestion rates of food of the population of Brodokalmak ( $\text{kg y}^{-1}$ ).

Food	Ingestion rate, $\text{kg y}^{-1}$					
	Adult		Children		Infants	
	Average	Most exposed group	Average	Most exposed group	Average	Most exposed group
Domestic vegetables	49	190	23	76	12	37
Potatoes	91	250	73	180	39	91
Root vegetables	31	160	20	86	9.2	51
Meat	14	63	11	37	7.4	24
Milk	75	330	79	480	170	370
Milk products	25	93	18	54	8.3	25
Chicken	11	42	6.1	26	4.5	20
Eggs	9.5	34	12	28	5.8	12
Bread	130	270	91	190	26	110
Cereals	17	60	17	37	10	67
Water	820	1200	550	960	170	570
Freshwater fish	24	100	16	34	4.2	16

Table 3.2. Annual occupancy rates of the population of Brodokalmak ( $\text{h y}^{-1}$ ).

Location	Occupancy rate, $\text{h y}^{-1}$					
	Adult		Children		Infants	
	Average	Most exposed group	Average	Most exposed group	Average	Most exposed group
Bridge	25	100	25	100	12.5	50
Flood plain	50	250	37.5	150	25	125

Radionuclide concentrations in foods and other environmental materials were extensively measured by IPAE in the Brodokalmak area in 1996 and 1997. The monitoring programme focused on  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ , considered to be the most radiologically important radionuclides, because of their mobility in the environment. The methods used to carry out the measurements are described elsewhere [Trapeznikov 1993]. A summary of the measurements is given in Table 3.3; when more than one measurement was taken the range of the observed value and the mean value are given. Vegetables used in the measurements were collected directly from the kitchen gardens of habitations in Brodokalmak, washed and prepared as for consumption before being analysed. All observations of  $^{137}\text{Cs}$  in vegetables were below limits of detection. Samples of freshwater fish were taken from both the Techa river and two lakes (Shablisk and Miassovo). These two lakes are situated at some distance from Brodokalmak and are not generally used by people living in the village who prefer to obtain their fish from other lakes closer to the village. However, the concentrations observed in the two lakes are considered to be typical of the lakes in the Brodokalmak area and were therefore included in the assessment. Radionuclide concentrations in fish caught in the Techa river are an order of magnitude higher than those measured in fish taken from the lakes. Samples of water were taken from two artesian wells commonly used by the population for drinking water and from the Techa river. Radionuclide concentrations in the river water are about two orders of magnitude higher than those observed in water taken from the wells.

Table 3.3. Concentrations of  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$  in foods and environmental materials measured in the Brodokalmak area ( $\text{Bq kg}^{-1}$ ).

Material	Number of samples	Radionuclide concentrations, $\text{Bq kg}^{-1}$			
		$^{90}\text{Sr}$		$^{137}\text{Cs}$	
		Range	Mean value	Range	Mean value
Soil (kitchen garden)	4	9.3 - 124	74	42.4 - 73.3	57
Pasture (on flood plain)	1		65		31
Water (artesian wells, drinking)	1/2		0.015	0.0036 - 0.0074	0.0055
Water (Techa river)	1		7.2		0.15
Milk (all cows)	7	1 - 5.6	3.6	1.8 - 230	55
Milk (cow grazing on non restricted pasture)	5	1 - 4.9	3.6	1.8 - 12	7.1
Milk (cows grazing on flood plain)	2	1.6 - 5.6	3.6	120 - 230	175
Potatoes	7	3.2 - 9.4	5.4		BLD <sup>1</sup>
Domestic vegetables (cabbage)	1		28.5		BLD
Root vegetables (beetroot)	1		5.2		BLD
Fish (Techa river)	1		340		580
Fish (Shablisch and Miassovo lakes)	2/3	41 - 48	45	38 - 92	58
Cow meat	1		0.6		0.6
Chicken	1		N/A <sup>2</sup>		N/A
Egg	1		0.6		2.8
Cereals (wheat)	1/2		1.4	1.26 - 1.32	1.3

Concentrations of  $^{137}\text{Cs}$  in milk obtained from cows grazing on pasture away from the river bank (mean value:  $7.1 \text{ Bq kg}^{-1}$ ) are more than an order of magnitude lower than those in milk taken from cows which also graze on the flood plains of the river (mean value:  $175 \text{ Bq kg}^{-1}$ ) despite the restrictions. The concentrations of  $^{90}\text{Sr}$  measured in the same milk samples did not show this effect. The observed difference in the behaviour of the two radionuclides in milk could be explained by low potassium contents of the soil of the Techa river banks; further investigations are being carried out to determine the concentrations of this element in the soils of the region to test this assumption. It is possible that the high  $^{137}\text{Cs}$  concentrations found in two milk samples were the result of a set of unusual conditions and such concentrations would not be found throughout the year. The doses estimated assuming no restrictions may, therefore, be over-estimated. Environmental transfer models were used to predict radionuclide concentrations which had not been measured or which were below limits of detection [Cabianca 1999(b)]. These models were also used to verify the consistency of the measurement data [Cabianca 1999(a) and (b)]. Table 3.4 gives the concentrations of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in various materials predicted by the models and the comparison with measurement data. Activity concentrations calculated using environmental models are generally in good agreement with the measurements. However, the model predictions for  $^{137}\text{Cs}$  in milk and beef based on activity concentrations found in grass from the river bank are lower than the measured concentrations in milk and higher than those measured in beef. There are a number of possible reasons for this discrepancy, for example, the effect of potassium discussed above, or the beef cattle may not have grazed on the river

<sup>1</sup> BLD: Below Limit of Detection

<sup>2</sup> N/A: measurement Not Available

bank pasture for the same period as the milk cows, or the grass sample may not be representative of the area grazed. The uncertainties associated with both measured and calculated activity concentrations are considered in the probabilistic dose assessment by assigning ranges to the concentrations as discussed by Cabianca et al [Cabianca 1999(b)].

Table 3.4. Concentrations of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in foods and environmental materials predicted by environmental models ( $\text{Bq kg}^{-1}$ ).

Material	Radionuclide concentrations, $\text{Bq kg}^{\text{B1}}$			
	$^{90}\text{Sr}$		$^{137}\text{Cs}$	
	Measured	Model prediction	Measured	Model prediction
Potatoes	3.2 - 9.4	3.7	BLD	0.4
Cabbage (domestic vegetables)	28.5	22.3	BLD	0.4
Beetroot (root vegetables)	5.2	7.4	BLD	0.28
Chicken (from eggs)	N/A	0.08	N/A	25
Chicken (from wheat)	N/A	0.0056	N/A	0.58
Milk (cows grazing on flood plain)	1.6 - 5.6	1.1 - 13	120 - 230	10 - 56
Cow meat (from grass on Techa river banks)	0.6	0.2 - 1.9	0.6	60 - 340
Cow meat (from meat of cows grazing on unrestricted pasture)	0.6	0.6	0.6	15

Measurements of gamma dose rates were made by IPAE on the banks of the Techa river, the bridge over the river and in the village. Gamma dose rates measured in the village ranged from 70 to 110  $\text{nSv h}^{-1}$ . These observations are consistent with background radiation levels in the region and were therefore taken to be background. Measurements on the river banks ranged from 70 to 1400  $\text{nSv h}^{-1}$ . A summary of the gamma dose rates measured in Brodokalmak is given in Table 3.5. These measurements were also used to determine distributions of the external doses for use in the probabilistic assessment [Cabianca 1999(b)].

Table 3.5. Gamma dose rates measured in the Brodokalmak area ( $\text{nSv h}^{-1}$ )

Location	Gamma dose rate, $\text{nSv h}^{-1}$	
	Range	Mean value/mid point
Village	70 - 110	90
Bridge	280 - 300	290
Techa river banks	70 - 1400	520

The dose coefficients for the three age groups for ingestion of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  were taken from ICRP Publication 72 [ICRP 1996]. Distributions of the dose coefficients were also required for the probabilistic assessment. These were based on ranges of dose coefficients given by Khursheed and Phipps (1998) with additional distributions being obtained by scaling [Cabianca 1999(b)].

### 3.3.1.2 Results for Brodokalmak

#### Current doses

Doses to individuals living in Brodokalmak due to the Techa river contamination calculated in the assessment are reported in Table 6. As discussed earlier these are the sum of the annual effective dose from external irradiation and the committed effective dose from ingestion in a year. It is unlikely that individuals in the most exposed groups consume all their foods at the highest intake rates and, therefore, doses due to ingestion of the two foods which give the highest doses were summed with doses from all other foods consumed at average intake rates. The doses given in Table 6 are deterministic, calculated using 'best estimate' parameter values. Percentage breakdown of the doses by exposure pathway and radionuclide were also determined and are given in Cabianca 1999(a) and 1999(b).

Table 3.6. Additional current annual doses received by the population of Brodokalmak from contamination of the Techa river ( $\text{mSv y}^{-1}$ )#.

Age group	Dose, $\text{mSv y}^{-1}$			
	Scenario 1		Scenario 2	
	Average	Most exposed group	Average	Most exposed group
Adult	0.14	0.43	0.92	3
Children	0.17	0.39	0.95	2.3
Infant	0.13	0.26	0.72	1.6

#### Deterministic results for scenario 1

All doses calculated assuming that restrictions are still in place are lower than the annual effective dose limit for members of the public of 1 mSv recommended by ICRP and the annual whole body dose limit of 5 mSv imposed by current Russian legislation [NRS 1987]. In both cases the limits apply to the additional dose over and above that incurred by natural background radiation; furthermore ICRP's dose limit applies to the dose that results from practices that are subject to its system of control, whereas the doses in the present situation arise from practices that were conducted under less stringent requirements. The Russian general dose limits for the population will change in year 2000 and will then include an effective dose limit of 1  $\text{mSv y}^{-1}$  [NRS 1996]. For average consumers annual doses vary between 0.13  $\text{mSv y}^{-1}$  and 0.17  $\text{mSv y}^{-1}$  for infants and children, respectively, while annual doses to individuals in the most exposed group range from 0.26  $\text{mSv y}^{-1}$  to 0.43  $\text{mSv y}^{-1}$ . The latter doses would increase by between 0.11 and 0.14 mSv, if individuals were assumed to eat all their foods at the highest intake rate. The most significant pathways for this scenario are ingestion of fish, milk and domestic vegetables. The contribution of ingestion of milk to the total dose is highest for infants (46 to 49%) while the contribution of ingestion of fish to the total dose is highest for adults (34 to 48%). Ingestion of domestic vegetables accounts for between 18% and 35% of the total doses. Potatoes form an important part of the diet of people living in Brodokalmak and this is reflected in the contribution of ingestion of this food to the total doses (up to 14%). External exposure is not an important pathway for this scenario, accounting for less than 6% of the total dose. Strontium-90 is the more important of the two radionuclides considered, contributing between 72% and 85% of the total dose.

# These are the doses received due to the contamination of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  and are in addition to those received due to other sources, e.g. natural background radiation.

### **Deterministic Results for scenario 2**

The annual doses to individuals in the most exposed group calculated assuming that restrictions are lifted were between 1.6 mSv  $y^{-1}$  and 3.0 mSv  $y^{-1}$ . These doses are significantly higher than the annual effective dose limit recommended by ICRP (1 mSv  $y^{-1}$ ) and the effective dose limit that will come into force in Russia in year 2000. The doses are within the range of doses that people receive from natural radiation (2-10 mSv  $y^{-1}$ ) [Pavlenko 1995]. Annual doses to average consumers range between 0.72 mSv  $y^{-1}$  and 0.95 mSv  $y^{-1}$ . The two most significant exposure pathways for this scenario are ingestion of fish and ingestion of milk. This is a result of the high concentrations of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  measured in fish caught in the Techa river and of  $^{137}\text{Cs}$  in milk from cows which also graze on the flood plains. Ingestion of drinking water is a significant pathway for this scenario, contributing up to 25% to the total dose. This is due to the high concentration of  $^{90}\text{Sr}$  in the water of the Techa river and the assumption made in the calculations that the water used for human consumption is extracted directly from the Techa river. The contributions of the two radionuclides included in the assessment are similar. Doses due to ingestion of  $^{90}\text{Sr}$  account for between 39% and 69%, while the contribution of  $^{137}\text{Cs}$  varies between 28% and 58%. The percentage contribution of external exposure to doses calculated for this scenario is similar to that of scenario 1 (less than 5%) despite the increased exposure during time spent on the river bank.

### **Probabilistic results**

The distributions for each of the parameter values were used to obtain distributions of the additional total doses to the population of Brodokalmak in a probabilistic assessment. A computer package, Crystal Ball, was used to calculate the distribution using a Latin Hypercube sampling technique. The values of the main parameters of the distributions of doses calculated for this assessment are given in Table 3.7. The statistically significant parameters reported are the mean value, the standard deviation, the median, the 5<sup>th</sup> and 95<sup>th</sup> percentiles. Also given are the percentages of the distributions exceeding an annual dose of 1 mSv. All the distributions are quite narrow, with 90% of the doses calculated within a factor of between 1.7 and 2.3 of the median value. The mean and the median values are close to the doses calculated in the standard assessment. The distributions of doses calculated assuming that restrictions are in place indicate that none of the individuals in the average group and a very small percentage of the individuals in the most exposed group (less than 2% for all age groups) are likely to exceed a dose of 1 mSv. If restrictions were to be lifted the probabilistic assessment shows that almost all of the individuals in the most exposed group (more than 99% of adults and children and about 94% of infants) would receive a dose higher than 1 mSv, while the probability of average consumers exceeding this dose would be significant for adults (29%) and children (52%) while smaller for infants (5.3%).

Table 3.7. Main parameter values of distribution of current annual doses received by the population of Brodokalmak ( $mSv y^{-1}$ ).

Scenario	Group	Age group	Dose, $mSv y^{-1}$					Percentage exceeding 1 mSv
			Mean	Median	Standard deviation	5 <sup>th</sup> percentile	95 <sup>th</sup> percentile	
Restrictions in place	Average	Adults	0.15	0.14	0.059	0.074	0.26	0.0%
		Children	0.21	0.19	0.083	0.10	0.36	0.0%
		Infants	0.10	0.090	0.046	0.045	0.19	0.0%
	Most Exposed	Adults	0.43	0.39	0.17	0.22	0.75	1.4%
		Children	0.48	0.44	0.19	0.25	0.84	1.9%
No restrictions	Average	Infants	0.30	0.27	0.14	0.14	0.56	0.2%
		Adults	0.87	0.80	0.35	0.42	1.5	29.1%
		Children	1.1	1.0	0.46	0.52	2.0	51.8%
	Most Exposed	Infants	0.51	0.45	0.26	0.21	1.0	5.3%
		Adults	2.6	2.5	0.93	1.5	4.3	99.8%
		Children	3.1	2.9	1.0	1.7	5.0	99.9%
		Infants	1.9	1.7	0.81	1.0	3.5	93.6%

### Future doses

Doses to the people of Brodokalmak in the future are likely to remain of the same order as the current doses. There is a relatively constant source of activity into the river and the situation can be regarded as approximately in equilibrium with any losses balanced by inputs from the MAYAK site. If there are no changes to this situation then the doses would eventually decline due to the radioactive decay of the main radionuclides of interest,  $^{90}Sr$  and  $^{137}Cs$ . However, as discussed earlier in this report, it is known that there are considerable quantities of radioactivity on the MAYAK site, notably in the Asanov swamp and in the various reservoirs on the site. There is, therefore, the possibility that levels of radioactivity could rise in the future, for example due to major flooding leading to greater than normal quantities of radioactivity being released from the Asanov swamp. There is also the possibility of a dam failure leading to the release of the significant quantities of radionuclides held in the reservoir into the Techa river. In this assessment, therefore, future doses to the population of Brodokalmak have been estimated assuming an enhanced release.

A representative computer model of the Techa river between the MAYAK plant and Brodokalmak was produced in order to model possible future radionuclide release scenarios. A compartmental model of the Techa river was derived using information on flow rates and other characteristics of the Techa river taken from Trapeznikov (1993). The model consists of four sections each containing a water column and bed sediment compartment [Cabanca 1999(b)] and is based on the model described in Simmonds (1995). Large variations in the flow rates between years were accounted for in the model by using annual measured and interpolated flow rates [NRPA 1997]. The source term for  $^{90}Sr$  discharges to the river was based on estimated discharges of radionuclides from the Asanov swamps and the bypass channels [NRPA 1997]. An estimate of 6% was given for the percentage of the  $^{137}Cs$  source term compared to that of  $^{90}Sr$ . Following implementation, the model for  $^{90}Sr$  and  $^{137}Cs$  was calibrated using available measured river water and bed sediment activity concentrations [Cabanca 1999(b), NRPA 1997].

Two options were considered to represent possible future events in the MAYAK region. A breaching of the final reservoir dam (reservoir 11) with the entire radionuclide inventory and water volume flowing down the Techa valley was chosen as the first event. The second event assumed that an increase in the spring floods led to increased runoff of 10% of the estimated radionuclide in-



ventories [NRPA 1997] from the upper reaches of the river with the annual flow rates kept the same as previous years. The doses resulting from the future scenarios have been calculated by using the ratios of radionuclide activity concentrations in river water and bed sediment predicted in the river model and those measured at Brodokalmak. The ratios have been used to scale the doses calculated in the assessment of doses for Brodokalmak assuming that scenario 2 (no restrictions in place) was being followed. The activity concentrations of meat, milk and milk products have been multiplied by the ratio of bed sediment activity concentrations calculated after and prior to the event. The activity concentration in the drinking water has been calculated by scaling the original measured activity concentration by the ratio of the modelled river water activity concentrations following the event and prior to the event. The activity concentrations in the fish caught in the river have been calculated by multiplying the scaled river water activity concentrations by freshwater fish equilibrium transfer factors. The doses have then been calculated using the same methodology as for the standard Brodokalmak dose assessment; no changes in habits were assumed. Calculated activity concentrations in fish are in the range of  $10^5$  Bq kg<sup>-1</sup> for both <sup>137</sup>Cs and <sup>90</sup>Sr, similar to the activity concentrations measured in fish taken from reservoir 11 [NRPA 1997].

The breaching of the reservoir 11 dam could result in doses in the order of several hundred millisieverts to a few sieverts for average and most exposed individuals respectively, assuming that no restrictions were introduced. The radionuclide of most importance for both sets of individuals is <sup>137</sup>Cs. The increased radionuclide runoff from the swamp could result in doses in the range of a few tens of millisieverts to around one hundred millisieverts for average and most exposed individuals, respectively. Caesium-137 contributes the majority to the total dose. Although the estimated doses are very approximate it is seen that future events could raise the doses received by over one and up to three orders of magnitude over the doses predicted in the standard dose assessment.

### 3.3.1.3 Estimated doses for Muslyumovo

Detailed measurements were not available for the village of Muslyumovo and therefore the Techa river model described above was used to estimate current doses received by adults in Muslyumovo, supplemented by additional published material [NRPA 1997]. For scenario 1, it was assumed that activity concentrations in foods were the same as at Brodokalmak, however measured gamma dose rates at Muslyumovo were used. For scenario 2, the Muslyumovo gamma dose rates were used and activity concentrations of several of the foodstuffs were scaled using the river model predictions. The activity concentrations of milk, milk products and meat were scaled by the predicted bed sediment activity concentration ratio between Muslyumovo and Brodokalmak. For drinking water, the activity concentrations were scaled by the ratio of the predicted river water activity concentrations at Muslyumovo compared with Brodokalmak. The scaled water activity concentrations were used to estimate the activity in fish caught in that section of the river by using equilibrium concentration factors [Simmonds 1995, NRPA 1997].

All doses presented here are approximate due to the lack of data. There is therefore a large uncertainty and the estimates should be used only as a guide to the actual doses received. The current doses estimated to be received by adults in Muslyumovo for scenario 1 were 0.2 mSv y<sup>-1</sup> for average individuals, whereas the most exposed individual consuming the two highest-contributing foodstuffs at critical rates and the remainder at average rates, received a dose of 0.6 mSv y<sup>-1</sup>. The critical pathway for both individuals is external dose, repre-

senting 29% and 36% of the dose received by the average and most exposed group, respectively. Fish and domestic vegetable consumption make a significant contribution to the dose of the most exposed individuals, 32% and 24%, respectively. The more important radionuclide considered is  $^{90}\text{Sr}$  for both cases, comprising 53% and 50% of the dose for average and most exposed adults, respectively. All the doses estimated for scenario 1 are lower than the annual effective dose limit for members of the public of 1 mSv recommended by ICRP [ICRP 1990].

Estimated doses received by adults in Muslyumovo using scenario 2 were much greater. An average adult was predicted to receive a dose of around 2 mSv  $\text{y}^{-1}$ . The most exposed individual is expected to receive a dose in the region of 6 mSv  $\text{y}^{-1}$  for consumption of two foodstuffs at critical rates and all foodstuffs at critical rates. The critical pathway for both individuals is fish consumption, comprising 51% and 64% of the total dose received by the average and most exposed group, respectively. The contribution from the two radionuclides is approximately equal for the average and most exposed groups. Both doses, assuming that no restrictions are in place, are in excess of the annual effective dose limit for members of the public of 1 mSv recommended by ICRP [ICRP 1990].

#### 3.3.1.4 Discussion of doses for Techa river populations

Current additional doses have been calculated for the people of Brodokalmak for two scenarios, assuming that the restrictions are followed completely and assuming that no restrictions are in place. In reality the situation is somewhere in between, with some restrictions being followed and some being ignored on occasion. For example, it is known that some cows graze on the river flood plain sometimes but many do not. Also, people may spend some time on the river banks and may fish but such occasions are likely to be limited and it is unlikely that all of their fish would be obtained from the river. It is unlikely that people drink any water directly from the river other than inadvertently while swimming. Such amounts and the resulting doses are very small. For the majority of people in Brodokalmak their doses are likely to be as assessed for Scenario 1. However, there may be individuals whose doses could approach those for scenario 2 depending on the extent to which they ignore the restrictions.

The people of Brodokalmak and Muslyumovo may be exposed through additional routes than those considered here. Some other pathways have been considered for Brodokalmak, notably those from wild foods such as mushrooms and berries, but the contribution to the total dose is insignificant as the river is the primary source of contamination. The possibility of cattle drinking the river water and people swimming in the water have also been considered, but again these pathways do not make a significant contribution to the total dose [Cabanca 1999(b)]. There is the possibility that wild game birds (ducks, geese etc) could live in the area and be eaten by the local population. It is possible that this pathway could contribute to the doses received by the population of the two villages and this requires further investigation. There will also be additional radiation doses due to other radionuclides in the area, for example  $^{239}\text{Pu}$ , but these doses are thought to be small compared to those from  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ .

The doses to the populations of Brodokalmak and Muslyumovo estimated here may be compared with those published elsewhere, for example see Kryshev et al [Kryshev 1998(a) and (b)]. The estimated doses are of the same order as the annual doses to the population along the Techa river calculated by Kryshev et al [Kryshev 1998(a)] for 1992 (0.1 to 2 mSv  $\text{y}^{-1}$ ). A second assessment by Kryshev et al [Kryshev 1998(b)] calculated doses for residents of the Techa river of between 0.08 and 0.7 mSv  $\text{y}^{-1}$  with an average value of 0.2 mSv  $\text{y}^{-1}$ .

These doses are lower than those calculated in the SUCON project. This is due to the use of a smaller ingestion rate of fish and lower concentrations of  $^{137}\text{Cs}$  in milk and fish used by Kryshev than those found here.

Future doses to the population are likely to remain of the same order as those estimated provided that there is no increase in the discharges to the Techa river. Estimates of possible future doses have been made assuming that there are increases in the discharges either resulting from a reservoir dam failing or from severe flooding. These indicate that radiation doses would rise significantly if no measures were taken to reduce exposures if such events were to occur.

### **3.3.2 The East Urals Trace**

#### **3.3.2.1 Dose Assessment methodology**

The village of Bagaryak is located about 70 km from the MAYAK plant and is on the edge of the east Urals trace area, ie the area affected by the Kyshtym accidental release in 1957. As far as is known there are no special measures in force at Bagarak which affect radiation doses. A similar deterministic dose assessment was carried out for Bagarak as for the village of Brodokalmak and outlined in Section 3.1. The doses estimated were the sum of the committed effective doses due to the ingestion of radionuclides in a year and the external effective radiation dose received in a year as defined by ICRP [ICRP 1990] for comparison with the annual dose limits.

The Institute of Plant and Animal Ecology (IPAE) carried out a programme of measurements of foods obtained from Bagarak in 1997 and 1998 to determine concentrations of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$ . These measurement data are summarised in Table 3.8 and in the assessment they were supplemented by additional data from environmental modelling [Cabianca 1999(b)]. The assessment considered the intakes of a range of terrestrial foods including wild foods (berries and mushrooms) [Cabianca 1999(b)]. IPAE also obtained information on the dietary intakes of the local population. As previously, doses were estimated for average individuals and a most exposed group, with higher food intakes and who spent longer outdoors, and three age groups were considered. Doses to infants and children were estimated in a similar way to those of adults, however detailed intake rate data were not available for Bagaryak, consequently the ingestion rates from Brodokalmak were used. The available information for children from Bagarak shows that the intakes are likely to be very similar. Adult members of the most exposed group were assumed to be farmers and as a result their outdoor occupancy was taken to be higher than that for the average members of the community at 2000 hours a year. That value equates to spending approximately half of every day outside for the six months of the year that climatic conditions allow. Average adults, children and infants were assumed to spend 900 hours a year outside, approximately 20% of the six months. All members of the local community were assumed to obtain their food from local sources.

Table 3.8. Activity concentrations of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in environmental materials measured in the Bagaryak area ( $\text{Bq kg}^{-1}$ )

Material	Number of samples	Radionuclide concentration, $\text{Bq kg}^{-1}$			
		$^{90}\text{Sr}$		$^{137}\text{Cs}$	
		Range	Mean value	Range	Mean value
Soil (forest)	2	14.1 - 38.7	26.4	17.0 - 52.7	34.8
Soil (floodplain)	2	35 - 59.7	47.4	65.3 - 71.0	68.2
Water (well)	2	0.025 - 0.14	0.083	0.098 - 0.2	0.15
Water (drill hole)	2	0.014 - 0.029	0.022	0.015 - 0.017	0.016
Water (lake Shablisch)	1		0.42		0.042
Milk	6	0.63 - 3.37	1.28	0.14 - 1.16	0.49
Potatoes	3	1.31 - 2.14	1.59	0.12 - 1.07	0.47
Carrots	2	0.92 - 2.41	1.67	0.21 - 1.63	0.92
Cabbage	1		0.41		0.43
Cucumber	1		2.88		0.43
Eggs	1		5.2		0.22

### 3.3.2.2 Results for Bagyarak

Current doses to average and the most exposed adults in Bagaryak are given in Table 3.9. Doses in Bagaryak are lower than those in Brodokalmak at approximately  $0.3 \text{ mSv y}^{-1}$  for the most exposed group and an average dose of  $0.1 \text{ mSv y}^{-1}$ . Table 3.9 also shows the contribution of the doses from the different exposure pathways considered and for the two radionuclides. For both the most exposed and average adults the consumption of freshwater fish is estimated to give about 50% of the dose, with around a further 40% coming from the consumption of terrestrial foods. The remaining doses for the most exposed group are around 4% from external exposure above contaminated soil and approximately 2% from the ingestion of drinking water. The inhalation of resuspended material makes a negligible contribution to the dose, even when enhanced resuspension due to mechanical activity such as ploughing or digging is considered. The contributions to the remaining dose to average members of the community are similar with around 6% from external exposure and 4% from drinking water.

Table 3.9. Current annual doses to adults in the village of Bagaryak#.

Pathway	Dose, $\text{mSv y}^{-1}$			
	Average		Most exposed group	
	$^{90}\text{Sr}$	$^{137}\text{Cs}$	$^{90}\text{Sr}$	$^{137}\text{Cs}$
Terrestrial foods	0.019	0.019	0.069	0.06
Freshwater fish	0.02	0.029	0.065	0.093
Drinking water	0.0022	0.0017	0.0034	0.0026
External	$7.5 \cdot 10^{-11}$	0.0056	$1.7 \cdot 10^{-10}$	0.012
Total	0.042	0.055	0.14	0.17
Grand Total	0.097		0.3	

Doses to children and infants in Bagaryak are given in Tables 3.10 and 3.11, in both cases the doses are lower than the corresponding doses to adults. The dose to children thought to be the most exposed were calculated to be around 0.2

# These are the doses received due to the contamination of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  and are in addition to those received due to other sources, e.g. natural background radiation.

mSv y<sup>-1</sup>, with just over 50% of that dose from the consumption of terrestrial foods. Average doses to children are lower at around 0.08 mSv y<sup>-1</sup> and the pathway breakdown is similar to that for children in the most exposed group. Those infants thought to be the most exposed receive a dose of around 0.17 mSv y<sup>-1</sup>, whilst the average dose to infants is approximately 0.05 mSv y<sup>-1</sup>.

Table 3.10. Current annual doses to children in the village of Bagaryak<sup>#</sup>.

Pathway	Dose, mSv y <sup>-1</sup>			
	Average		Most exposed group	
	<sup>90</sup> Sr	<sup>137</sup> Cs	<sup>90</sup> Sr	<sup>137</sup> Cs
Terrestrial foods	0.03	0.0018	0.097	0.0066
Freshwater fish	0.024	0.013	0.051	0.029
Drinking water	0.0027	0.00083	0.0047	0.0014
External	7.5 10 <sup>-11</sup>	0.0056	7.5 10 <sup>-11</sup>	0.0056
Total	0.057	0.022	0.15	0.042
Grand Total	0.079		0.19	

Table 3.11. Current annual doses to infants in the village of Bagaryak<sup>#</sup>.

Pathway	Dose, mSv y <sup>-1</sup>			
	Average		Most exposed group	
	<sup>90</sup> Sr	<sup>137</sup> Cs	<sup>90</sup> Sr	<sup>137</sup> Cs
Terrestrial foods	0.032	0.0022	0.11	0.0060
Freshwater fish	0.0077	0.0046	0.029	0.017
Drinking water	0.001	0.00033	0.0034	0.0011
External	7.5 10 <sup>-11</sup>	0.0056	7.5 10 <sup>-11</sup>	0.0056
Total	0.041	0.013	0.14	0.03
Grand Total	0.054		0.17	

In the future, doses are likely to decrease through the processes of radioactive decay and migration of radionuclides into deeper layers of the soil. A validation exercise was carried out to ensure that the model for the migration of radionuclides was suitable for use in predicting changes in activity concentrations in the environment with time [Busby 1999]. Results [Cabianca 1999(b)] suggest that doses from both <sup>137</sup>Cs and <sup>90</sup>Sr are likely to fall to around 75% of their current value in ten years, falling to approximately 40% in thirty years. Doses due to the consumption of freshwater fish constitute a considerable fraction of the dose received by the inhabitants of Bagaryak and it is difficult to predict how this dose will change in the future apart from through radioactive decay. If there are no further releases to the environment affecting Bagaryak then doses from all exposure pathways will decline gradually in the future due to a combination of radioactive decay and physical processes.

### **Discussion of doses for the east Urals trace**

The results presented here are based on a limited set of measurements and it has not been possible to find other published assessments for comparison, although the results are consistent with predictions of environmental modelling [Cabi-anca 1999(b)]. Doses are likely to be similar in other settlements in the region with one of the most important factors being where people obtain their fish from. Assuming a contamination level in the restricted area of  $37 \text{ kBq m}^{-2}$  it has been estimated that current radiation doses would be between  $0.5$  and  $1 \text{ mSv y}^{-1}$ . As discussed above for Brodokalmak there are uncertainties associated with any dose assessment and in this case it has not been possible to quantify these uncertainties, although they are likely to be of a similar order as those given in section 3.3.1.2.

## **3.4 Strategies for Remedial Measures**

The objectives for this part of the project (WP4) were to consider strategies for the reduction of the radiation doses received by critical groups in the Southern Urals and to consider strategies for the reclamation of contaminated land areas. These overall objectives were met by considering the measures already in place in the region, by reviewing remediation measures taken elsewhere and by considering the implications of different options, including the effect on future radiation doses.

The remediation measures that have been carried out in the Southern Urals to reduce the radiation exposures resulting from the various sources of contamination have been reviewed [Bexon 1999(a) and (b)]. A review has also been made of information relating to remediation measures that have been carried out or considered elsewhere [Bexon 1998, 1999(b)]. In addition the radiation dose assessment carried out for the region has considered the impact of any remedial measures in place. Based on this work recommendations are made on possible strategies to reduce the radiological consequences of the levels of contamination present in the region.

### **3.4.1 Remedial techniques carried out in the Southern Urals area**

As previously discussed significant quantities of radioactive waste was discharged to the Techa river between about 1948 and 1951. Subsequently waste was discharged into Lake Karachay and Reservoir 17. The Techa River releases led to high levels of contamination in the region and remedial measures started in 1951. Firstly, a series of dams were constructed to create an enclosed series of lakes isolated from the rest of the Techa river with canals to the north and south to divert the uncontaminated river water. By 1960 about 6300 people were evacuated from 22 villages in the upper reaches of the Techa river. An exclusion zone was created around the MAYAK site including the lakes, reservoirs and the Assanov marshes where significant quantities of radionuclides are retained. Further long-term measures were also introduced to reduce the exposure of people living in settlements on the banks of the Techa river. Floodplains were fenced off and the access of people and livestock to these areas was restricted. Restrictions on drinking water and fishing in the river were also introduced. Construction of wells and pipelines to provide uncontaminated drinking water started in 1953 and by 1954 all inhabitants and cattle were provided with water from other sources. However, some consumption of river water is thought to have continued until 1956 when further measures were taken to enforce the restrictions. These long-term measures are still in place although there is evidence that villagers do not always follow the restrictions, sometimes fishing in

the river or letting their animals graze on the floodplains. Plans were made for further remediation measures but they have not yet been implemented. The priority seems to be to prevent further contamination reaching the occupied areas by taking various remedial measures within the restricted area. However, consideration is also being given to reducing exposures on the river banks downstream through measures such as removal of the top layers of sediment or by shielding the contaminated sediment with crushed rocks.

Following the 1957 accident at the MAYAK plant, the so called Kyshtym fire, radionuclides were deposited over a large area downwind. Some 1,100 people were evacuated in the first 7 to 10 days from the most affected area. A further 10,000 people were evacuated at varying times up to 670 days following the accident. Access was restricted for an area of about 20,000 hectares and 106,000 hectares of land were removed from agricultural production for a minimum period of 4 years. In the short term after the accident, the most severely contaminated animals (about 200 cattle and 300 sheep) were culled and buried. Measures, such as the provision of uncontaminated feed were taken to reduce the transfer of radionuclides to animal products and other terrestrial foods. Fishing was also forbidden in the two most affected lakes. Treatment of 20,000 hectares of the most affected land was carried out in 1958 and 1959 and mainly involved ploughing the surface layers. An additional deeper ploughing of 6,200 hectares to a depth of 50 cm was undertaken between 1960 and 1961. Another measure that has been reported in the open literature was the devegetation of contaminated forest areas between 1959 and 1963. This measure would have led to 50% of the contamination being removed, depending on the cover type and density. The exclusion area appears to have been reduced in recent years but there are still significant areas of land which are not occupied and where the only agricultural production is in specialised state farms for research purposes. It does not appear that any specific measures were taken following the resuspension of contaminated sediments from Lake Karachay and their subsequent distribution over a wide area of the Southern Urals.

The remediation measures carried out in the Southern Urals are discussed in more detail in Bexon 1999(a) and (b).

### **3.4.2 Remedial measures taken outside the Southern Urals area**

The remediation measures that are possible after an incident resulting in the radioactive contamination of land include both short term and longer term countermeasures, and also techniques which are applicable to severe cases of contamination or to small areas. All measures were included in the reviews carried out [Bexon 1998, 1999(b)] but were subdivided into those techniques which may be applicable to the Southern Urals contamination and those which are unlikely to be applicable, usually because the measures are only effective if undertaken within the first weeks or months of the contamination occurring. The exposure pathways the countermeasures are primarily effective against was also considered. Table 4.1 lists the countermeasures which have been proposed for mitigating the effects of contamination, including both decontamination techniques and agricultural countermeasures. The techniques in Table 4.1 are those which are potentially of use in reducing doses in the southern Urals area, and these are discussed in more detail below. Bexon 1998 and 1999(b) also include other measures which are not thought to be applicable to the Southern Urals, for completeness and to provide reassurance that no potentially useful countermeasure has been overlooked.

Table 4.1. Remediation measures potentially applicable to the Southern Urals

Remediation measure	Effective against internal or external exposure?	Any notes
Soil and/or turf removal and replacement	External Internal if agricultural land	Could be done in a limited area to provide clean grazing facilities
Access restrictions to most contaminated areas	External Internal if land used for agriculture or grazing	
Provision of clean food supplies	Internal	
Provision of clean drinking water	Internal	
Establishment of food supply system to encourage use of less contaminated produce	Internal	For example, provision of fish from less contaminated lakes
Food preparation	Internal	Ingestion doses may be reduced by careful preparation of food before consumption, especially vegetables
Continuous monitoring system	External and internal	To act as early warning of an increase in levels so rapid action can be taken
Enforcement of restriction countermeasures	External and internal	
Alternative land use		For example, establishment of alternative crops such as forestry, fibres, paper or fuel.
Application of soil treatments	Internal	Land improvement, application of inorganic fertilisers or liming
Phytoremediation	Mostly internal	The growth of crops such as sunflowers and spinach which absorb radionuclides from the soil. The crops are then disposed of.
Surface improvement	Internal	A combination of techniques including drainage, fertiliser and grass seeding. May be applicable to flood plains
Drainage improvement	Internal	May reduce root uptake by lowering the water table.
Application of chemicals to water bodies	Internal	To reduce uptake by fish, lime or potassium chloride may be added to water
Transfer of fish to uncontaminated tanks for short periods before consumption	Internal	To reduce concentrations in fish
Selection of crop varieties and species	Internal	To reduce uptake by roots.
Animal chemical treatments	Internal	Addition of binders to feed
Animal feed	Internal	Management of animal feeding regime, including clean feed.
Animal husbandry	Internal	Removal from contaminated areas to cleaner areas prior to slaughter.
Semi-natural ecosystem improvement	Internal	Improve relatively clean but poor areas to provide land for fattening prior to slaughter
Education of the local inhabitants	External and internal	Improved awareness of the contamination situation and ways to reduce exposure
Relocation	External and internal	Would only be justified if significant doses are still being received



A key factor in determining which countermeasures should be implemented in a contaminated area is the economic and other costs associated with each measure. Some of the measures listed above are have relatively small costs, for example, advice on food preparation in the home, and education of the local inhabitants. Some measures would have initial implementation costs but the longer term maintenance of the measures is comparatively cheap, for example the use of access restrictions to the most contaminated areas, and the establishment of food supply system to encourage use of less contaminated produce. The establishment of a continuous monitoring system would have initial costs but subsequent maintenance would be cheaper. Permanent relocation of the affected population has large costs, both economic and social, in the initial stages, but these decrease with time as the population settles in the new location.

Other measures are more expensive, both to implement and to maintain, such as the continual provision of clean food supplies and clean drinking water. Some countermeasures have very substantial costs associated with them, and would be more suited to application in small areas of high contamination. These would include the application of soil treatments, animal chemical treatments, and soil and/or turf removal and replacement.

### **3.4.3 Radiological implications of remedial measures in the Southern Urals**

As discussed earlier a dose assessment was carried out for the village of Brodokalmak on the Techa river which considered two possible scenarios. In the first scenario it was assumed that the restricted access to the river banks for both people and cattle was observed by the population. For the second scenario, doses were calculated assuming that the restrictions were lifted and people had free access to all areas in the village. Doses were calculated for three age groups (adults, children and infants) and two types of individuals: average consumers and users of the river banks, and individuals most likely to receive the highest dose. These individuals are referred to as the most exposed group. The exposure pathways identified as the most radiologically significant were ingestion of terrestrial and aquatic foods and exposure to gamma radiation deposited on the banks of the river. Doses due to ingestion of vegetables, chicken, eggs, bread and cereals remain unaffected by the lifting of the restrictions, because crops are not grown on the river flood plains. In addition to the standard dose assessment for the population of Brodokalmak as described earlier, a probabilistic assessment was also undertaken. For this assessment, ranges and distributions of the input parameters were defined and distribution of doses were then calculated using a spreadsheet program by sampling from the distributions of the input parameters. The assumptions and methodology adopted for the probabilistic assessment were the same as those used in the standard assessment.

To assess the efficacy that the imposition of restrictions to the Techa river could have in reducing the doses received by the population of Brodokalmak dose averted by the application of restriction to access to the Techa river have also been calculated [Bexon 1999(b)]. These doses were calculated by subtracting doses calculated for scenario 1 (restrictions in place) from the corresponding doses calculated for scenario 2 (restrictions lifted). For the most exposed group doses averted were calculated assuming that all the foods are consumed at higher than average rate, rather than only the two foods which give the highest doses. This different approach was necessary because the two foods which give the highest doses are not the same for the two scenarios. The averted doses for adults are given in Table 4.2 ranked in descending order, Bexon 1999(b) also gives averted doses for children and infants. For doses due to ingestion of foods the contribution of both radionuclides included in the assessment are presented separately. The imposition of a ban on consumption of fish caught in the Techa

river and drinking water abstracted from the river reduces doses to average adults and children by 0.5 mSv to 0.6 mSv and to the most exposed group by 1.2 mSv to 1.8 mSv. Doses saved by restricting access of cows to pastures on the flood plain are lower with the greatest saving being about 0.9 mSv for the most exposed group of adults. The reductions in doses to infants achieved by the imposition of restrictions are smaller but still significant. Larger reductions in doses received by infants are achieved by banning cows from the flood plain (0.37 mSv for average consumers and 0.79 for most exposed individuals). Doses averted by restricting access of people to the flood plain of the Techa river are lower. Doses saved are of the order of 0.02 mSv for average users of the river banks and between 0.06 mSv and 0.12 mSv for individuals in the most exposed group.

*Table 4.2. Dose to adults living in Brodokalmak averted by the application of restrictions.*

Rank	Average		Most exposed group	
	Nuclide, Pathway	Dose saved (mSv)	Nuclide, Pathway	Dose saved (mSv)
1	Sr-90, Fish	0.2	Sr-90, Fish	0.86
2	Sr-90, Water	0.17	Cs-137, Milk	0.72
3	Cs-137, Milk	0.16	Cs-137, Fish	0.7
4	Cs-137, Fish	0.16	Sr-90, Water	0.24
5	Cs-137, Milk products	0.055	Cs-137, Milk products	0.2
6	Total, External	0.024	Total, External	0.12
7	Cs-137, Meat	0.0025	Cs-137, Meat	0.012
8	Cs-137, Water	0.0016	Cs-137, Water	0.0023

### **3.4.4 Recommendations on possible remediation measures in the Southern Urals**

Based on the reviews described in Bexon 1998, 1999(a) and 1999(b), together with the dose assessment outlined above the following recommendations are made on possible measures that should be taken to reduce the impact of the radioactive contamination in the Southern Urals.

#### **Techa river area**

- Average doses are relatively low and the introduction of new countermeasures does not appear justified.
- The principal recommendation is the establishment of a continual monitoring system that will provide an alert if environmental levels of activity should for any reason increase
- An education programme to familiarise the local populations with the sources of local exposure, the magnitude of this, and the health impact may reduce anxiety.
- Advice on the best sources of foods (such as information on which lakes are less contaminated) may be helpful.
- Restrictions on the consumption by humans of fish and water from the Techa river, and the restriction of cows from grazing on the river flood plains are successful in terms of dose reduction. They should be continued and enforced.

- The doses that are averted by restricting access of people to the flood plain of the Techa river are low. It should be recognised that the occasional visit to the floodplains or the river bank for recreational purposes (rather than for fishing or for the grazing of animals) is not likely to give rise to substantial doses.
- Good contacts between the local inhabitants and those with responsibility for improving the situation in the area should be established.
- More intensive and expensive measures are not justified by the levels of dose.

It is understood that some of these measures are already underway in the region and these should be maintained.

### **Bagaryak**

- The current doses are low and do not require the introduction of restrictions.
- A population reassurance programme could reduce anxiety levels, if these exist.
- Advice on which lakes have the lowest levels of fish contamination could be beneficial.
- There is no indication that future doses in the area will give rise to any concern.
- The results of the assessment indicate that although the remediation decisions taken in the recent past in the vicinity of Bagaryak were appropriate, there are indications that these can now be relaxed.

### **Surrounding area**

Remedial measures are still required:

- In the region defined by the East Ural radioactive trail, somewhere between the MAYAK site and Bagaryak (it is known that restrictions are no longer required at Bagaryak, but it is not clear from the currently available data how far towards the site this situation extends).
- Along the banks of the Techa, between MAYAK and Muslyumovo.
- In the MAYAK area; in the absence of more information, it is not yet clear to what radius around the plant restrictions may still be required.

The nature of the remedial measures which may be appropriate in this area include:

- Alternative land use (for example, the growth of forestry, fibres or paper crops).
- Land improving soil treatments (including surface treatments and drainage).
- Phytoremediation (the growth of crops which will have a decontaminating effect such as sunflowers).

### **Other points**

Further measurements which would clarify the need for current and future restrictions include

- more data on the levels of activity in the fish actually consumed in the three villages
- more data on the levels in terrestrial foods in Bagaryak and Muslyumovo.

### 3.5 Global Impact Assessment

The major pathway for the global impact of the various radioactive releases from Mayak is the releases to the Techa river and hence to the Ob-river system. Information on the contamination of the Ob-river system outside the Techa-Iset rivers, in particular in the early years around 1950, are essentially nil. A major purpose of work package 5 has therefore been to measure samples of sediments from the Ob river system, because it is believed that such samples are suited for a retrospective mapping of the contamination of the river system in previous years.

The recipient of contamination carried out by the Ob-river is the Kara Sea in the Arctic Ocean. The Norwegians has in a joint international co-operation with the Russians carried out a number of studies in the Kara Sea primarily to study the radiological impact of the Russian dumping activities along Novaya Zemlya. These studies have, however, also dealt with the impact of run off to the Arctic Ocean with the Ob river.

#### 3.5.1 American, Danish-Russian studies in the Irtysh-Ob rivers

In cooperation with Woods Hole Oceanographic Institution (WHOI) a sediment core collected by WHOI from the Nentinsky sorlake in the Ob river system have been analysed for  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{239,240}\text{Pu}$ ,  $^{241}\text{Am}$  and excess  $^{210}\text{Pb}$ . The position of the sample was  $66^{\circ}32'.771\text{E}$ . The sample was collected 25 July, 1994. The core was cut in 1 cm thick slices and the results of the analysis are shown in Figure 5.1.

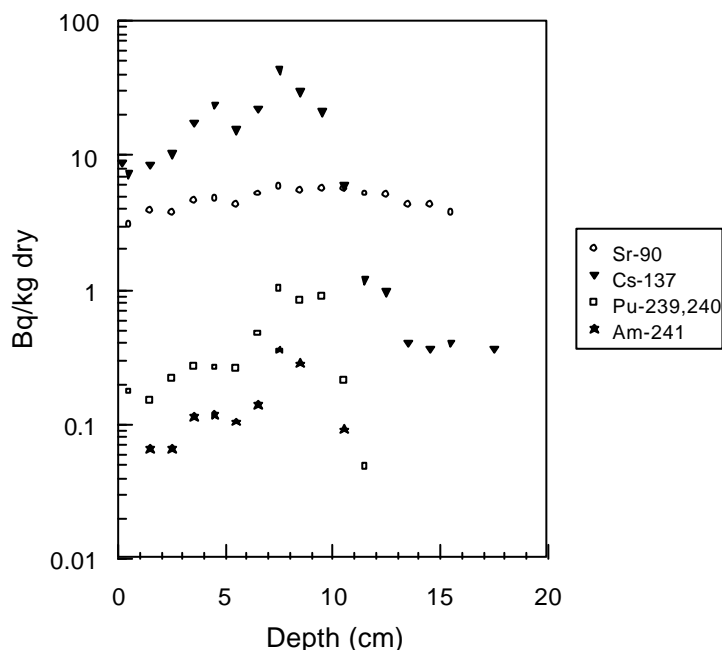


Figure 5.1. Radionuclide concentrations in Ob river sediments collected by WHOI in July 1994 from Nentinsky sorlake.

Neither in this sample nor in the many samples collected and analysed by WHOI (Panteleyev *et al*, 1995) was found any indication of radionuclides which could be attributed to the major releases from Mayak around 1950.

Mass spectrometric analysis for transuranics carried out by WHOI suggest, however, debris from other non fallout sources, perhaps Tomsk-7, in river sediments from the Ob-river system. (Sayles *et al*, 1997) (see also 3.6.7 in this report). The upper  $^{137}\text{Cs}$  peak at 4-5 cm in the sediment core may carry some activity from the Chernobyl event, but the dating of the core and the relative high  $^{90}\text{Sr}/^{137}\text{Cs}$  in the peak does not support this assumption. On the other hand the dating of river sediments may be difficult, because secondary depositions may occur downstreams a river, as discussed below for the Techa and Iset data from 1996. Furthermore the sedimentation rate may vary from one year to another.

### 3.5.2 Joint Russian-Danish-Ukrainian studies in the Techa-Iset-Tobol rivers.

These studies began in 1990 and samples of sorlake sediments and floodplain soils along the Techa-Iset and Tobol rivers have since then been collected by IPAE. (Trapeznikov *et al*, 1993, Aarkrog *et al*, 1997). The most recent results have been submitted for publication in Journal of Environmental Radioactivity and are summarized here.

Two sets of samples: floodplain soil sorlake sediments were in 1996 collected by Institute of Plant and Animal Ecology (Inst. name: Biophysical station IPAE in Zarechny) in Yeakaterinburg and analysed by Risø National Laboratory (RISØ) for  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ , Pu, Am and excess  $^{210}\text{Pb}$ . Most of the samples were also analysed by IPAE for  $^{137}\text{Cs}$  and a limited number of samples were analysed by Institute of Biology of the Southern Seas (IBSS) in Sevastopol, Ukraine.

Floodplain soil, sorlake sediments and river sediments (Nadirov Bridge) were all used together in the calculation of the radionuclide inventories in the river system. Figs. 5.2. and 5.4. show the calculated depositions of the  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and Pu in 1 m columns together with the fitted power functions. We assumed that all radioactive contamination of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  were present in the upper 100 cm layer of soils and sediments. The depositions below the sampling depths were calculated from the distributions in the upper layers, which in most cases showed an exponential decrease with depth. In a few cases especially for  $^{90}\text{Sr}$  this decrease was not evident and the calculation of the  $^{90}\text{Sr}$  inventories in the layers below the sampling depth are in such cases a guess, where we assume that the concentration in the deeper layers is the same as the mean of the measured upper layers.

As shown in Figs. 5.2.-5.4. the depositions of the radionuclides decrease after power functions ( $y=e^{ax^b}$ , see table 5.1) with distance from MAYAK. In case of  $^{90}\text{Sr}$  the regression was not significant, so we have for this radionuclide assumed a constant deposition density equal to the geometric mean of  $\text{kBq } ^{90}\text{Sr m}^{-2}$  in all 1 m columns ( $179 \text{ kBqm}^{-2}$ ) between Nadirov Bridge and Bolshie Pogrorelki, 49 and 310 km respectively from MAYAK. From 310 km and to the outlet of Ob (~2800 km from MAYAK) we assumed that the  $^{90}\text{Sr}$  deposit decreased after the power function shown in fig. 5.2. ( $^{90}\text{Sr}$ ), but this assumption is tentative.

The samples used in our inventory calculations were collected in the rivers and floodplain within a belt of approximately 50 m so we have assumed the samples to represent the contamination out to 25 m on each side from the middle of the riverbed. The contamination extends farther than that but decreases with the distance from the riverbed. The general spatial distribution of radioactive contamination levels in the flood plain has been assessed from gamma radiation dose rates for the Asanow Swamps over a distance of 40 km along the Techa river (NREG,1997). The  $^{137}\text{Cs}/^{90}\text{Sr}$  ratio in the floodplain was expressed by the power function:

$$^{137}\text{Cs}/^{90}\text{Sr} = 660.7 R^{-1.18} \quad (1)$$

where R is the distance from the riverbed in meters. From these Russian data we have calculated the following expressions for the horizontal distribution of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  as a function of the distance R from the river bed.

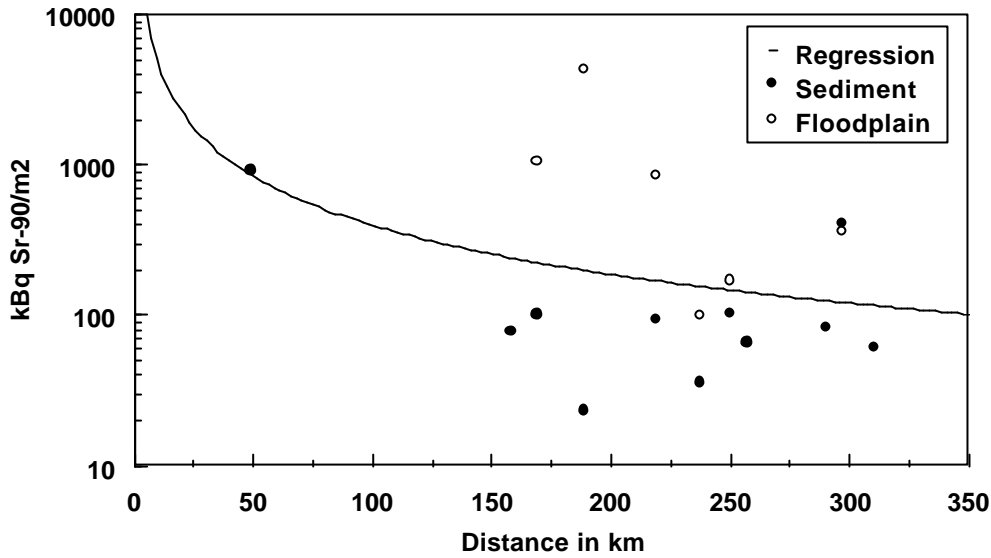


Figure 5.2. Cumulated Sr-90 in the 0-100 cm sediment and floodplain layer along the Techa and Iset rivers in 1996.

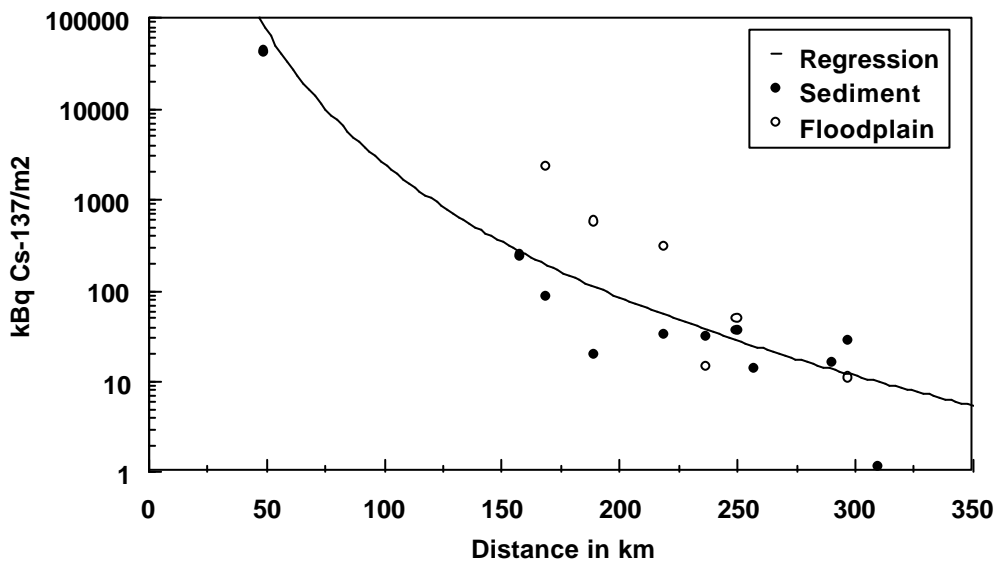


Figure 5.3. Cumulated Cs-137 in the 0-100 cm sediment and floodplain layer along the Techa and Iset rivers in 1996.

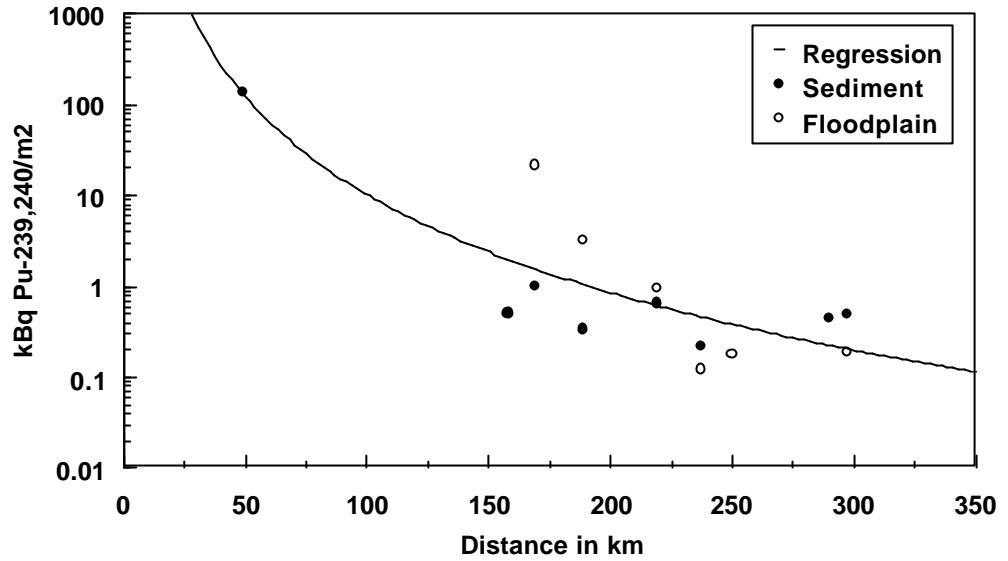


Figure 5.4. Cumulated Pu-239, 240 in the 0-100 cm sediment and floodplain layer along the Techa and Iset rivers in 1996.

$$\text{Bq } ^{90}\text{Sr m}^{-2} = A \cdot \frac{B \cdot e^{-0.0125R}}{660.7 R^{-1.18}} \quad (2)$$

$$\text{Bq } ^{137}\text{Cs m}^{-2} = B \cdot e^{-0.0125R} \quad (3)$$

$$\text{Bq } ^{239,240}\text{Pu m}^{-2} = C \cdot e^{-0.0125R} \quad (4)$$

We assume that the horizontal distribution of Pu follow that of  $^{137}\text{Cs}$  and that the deposition of  $^{137}\text{Cs}$  is proportional to the gamma-dose rate, which decreases exponentially with the distance from the river bed with a half distance of about  $55 \pm 10$  m (calculated from five transects in the floodplain (Fig. 5.7 in NREG, 1997)). From eqs. 2-4 it is calculated that in order to obtain the total inventories of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  in the floodplain the deposits found in the 50 m belt should be multiplied by 32, 3.8 and 3.8 respectively. We have in this calculation assumed that equations 2-4 are valid for the entire river system (A, B and C vary with the distance from MAYAK). This assumption may not be correct, but for  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  most of the activity is deposited in the upper reaches of the river system from where the equations were calculated, so for these radionuclides it has less implications if the assumption is wrong. In case of  $^{90}\text{Sr}$  the error may be greater because the  $^{90}\text{Sr}$  has moved further down-streams than  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$ .

The reservoirs built in the upper parts of the Techa river extend to Dam 11, situated 31 km from the original release point at MAYAK. From Dam 11 to Nadirov Bridge we have no samples but Russian data (NREG, 1997) are available from the Asanov Swamp from a location situated 7 km down streams from Dam 11. The levels of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  at this location (1200, 44000 and 74  $\text{kBq m}^{-2}$  respectively) are so close to ours obtained from Nadirov Bridge (920, 43000 and 137) that we for the first part of the river (18 km) after the reservoirs assume the depositions of the 3 radionuclides to be represented by the

means of these two sets of observations. The inventories are then calculated from these mean values by multiplication with the area of the 50 m belt (900 000 m<sup>2</sup>) and the above mentioned factors (3.8 for <sup>137</sup>Cs and <sup>239,240</sup>Pu and 32 for <sup>90</sup>Sr).

Table 5.1 Coefficients ( $\pm 1SD$ ) in the equation:  $y = e^a \cdot x^b$ , where  $y$  is kBq m<sup>2</sup> related to the distance  $x$  km from the original discharge point from MAYAK to the Techa river. (N: number of data).

Radionuclide	a	b	N
<sup>90</sup> Sr	11.0 $\pm$ 4.2	-(1.08 $\pm$ 0.79)	17
<sup>137</sup> Cs	30 $\pm$ 3.9	-(4.9 $\pm$ 0.72)	17
<sup>239,240</sup> Pu	18.9 $\pm$ 3.7	-(3.6 $\pm$ 0.70)	14

Table 5.2. Calculation of inventories of MAYAK derived <sup>90</sup>Sr, <sup>137</sup>Cs and <sup>239, 240</sup>Pu (TBq) in the Techa-Iset-Ob rivers and floodplain system. (cf. the text).

Part of the river (distance from original release point to the Techa river from MAYAK)	<sup>90</sup> Sr	<sup>137</sup> Cs	<sup>239,240</sup> Pu
Dam 11 – Nadirov (31 – 49 km)	31 a/	150 a/	0.36 a/
Nadirov – Bolshi Pogorelki (49 – 310 km)	76 b/	198 d/	0.47 d/
Bolshi Pogorelki – Guba Obskaya (310 – 2823 km)	~120 c/	~0 /d	~0 d/
Dam 11 – Muslymovo (31 – 78 km)	38 a,b/	315 a,d/	0.69 a,d/
Total river and floodplain beyond Dam 11 (31 – 2823 km)	0.2 PBq	0.35 PBq	0.8 TBq

### Examples of calculation

$$a/ \quad {}^{90}\text{Sr}: \quad \frac{920+1200}{2} \cdot 18 \cdot 50 \cdot 10 \cdot 10^{-6} = 31 \text{ TBq } {}^{90}\text{Sr}$$

$$b/ \quad {}^{90}\text{Sr}: \quad 179(310-49) \cdot 50 \cdot 32 \cdot 10^{-6} = 76 \text{ TBq } {}^{90}\text{Sr}$$

$$c/ \quad {}^{90}\text{Sr}: \quad \left( \int_{310}^{2823} 5.84 \cdot 10^4 \cdot x^{-1.08} dx \right) \cdot 50 \cdot 32 \cdot 10^{-6} = 120 \text{ TBq } {}^{90}\text{Sr}$$

$$d/ \quad {}^{137}\text{Cs}: \quad \left( \int_{49}^{310} 1.46 \cdot 10^{13} x^{-4.9} dx \right) \cdot 50 \cdot 3.8 \cdot 10^{-6} = 198 \text{ TBq } {}^{137}\text{Cs}$$

The uncertainty of the inventory estimates was obtained from a parameter uncertainty analysis carried out by the programme "Crystal Ball". We calculated 1SD ranges for the inventories between Nadirov Bridge and Bolski Pogorelki in table 5.2.: 51-107 TBq <sup>90</sup>Sr, 58-670 TBq <sup>137</sup>Cs and 0.13-1.67 TBq <sup>239,240</sup>Pu. This corresponds to uncertainty factors of 1.45, 3.4 and 3.6 for the 3 radionuclides respectively.



According to Russian estimates (NREG, 1997) 0.27% of the  $^{90}\text{Sr}$  in the Techa river system (river + floodplain) between Dam 11 and Muslymovo is to be found in the bottom deposits in the river. For  $^{137}\text{Cs}$  the percentage is 2.1. If we assume that this percentage is valid also for Pu and if we furthermore assume that all the percentages are valid for the entire Techa river, we may calculate by the methods shown above, how much the inventories in the river deposits between Nadirov and Zatechenskoe which were studied in 1990 (Trapeznikov *et al*, 1993) should be. We find 0.15 TBq  $^{90}\text{Sr}$ , 4 TBq  $^{137}\text{Cs}$  and 10 GBq  $^{239, 240}\text{Pu}$ . In 1990 we estimated these inventories to 0.3 TBq  $^{90}\text{Sr}$ , > 6 TBq  $^{137}\text{Cs}$  and 8 GBq  $^{239, 240}\text{Pu}$ . The agreement between the two estimates are surprisingly good and may be fortuitous, but suggests that the previous inventory estimate for the river deposits may be compatible with the present study.

The above-mentioned joint Norwegian-Russian study (NREG, 1997) has estimated the inventories of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the upper reaches of the Techa-river-floodplain between Dam 11 and Muslymovo, i.e. 31-78 km from MAYAK, to 36-44 TBq and 190-230 TBq respectively. These figures are comparable with those calculated in table 5.2. for this part of the river. The two estimates are not fully independent as we in our calculation has used one Russian data set (Asanov Swamp) and furthermore have calculated our horizontal distributions of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  in the floodplain from the Russians observations (eqvs. 2 and 3).

In a Russian study (Chesnokov *et al*, 1998 and Shcherbak, 1998) applying collimated scintillator detector techniques for the determination of the  $^{137}\text{Cs}$  contamination of the Techa river floodplain, five sites at the river floodplain was mapped: Muslymovo, Brodokalmak, Russkaja Techa, Zatechenskoye and Krasnoitsetskoye (Iset). The study covered about 68 km of the river system and the total number of measuring points were about 20000. This work is thus probably the most comprehensive investigation of the  $^{137}\text{Cs}$  contamination of the downstream parts of the Techa (Iset) river floodplain carried out so far. We may use the Russian inventories from this study to validate our model for  $^{137}\text{Cs}$  (Table 5.3.).

Table 5.3. Comparison of Chesnokov *et al* (1998) and Shcherbak (1998)'s  $^{137}\text{Cs}$  inventories with SUCON model calculations (see table 5.2.).

Location	Part of river (distance in km from MAYAK)	Chesnokov & Shcherbak Measurement (TBq)	SUCON model calculation (TBq)
Muslymovo	70-87	6.5	28
Brodokalmak	107.5-122.5	2.9	3.7
Russkaya Techa	134-160	1.24	1.99
Zatechenskoye	234-240	0.32	0.044
Krasnoitsetskoye	245-249	0.074	0.022

According to Table 5.3. the SUCON model seems to overpredict the levels in the upper parts and underpredict in the lower parts of the river system. About 99% of the  $^{137}\text{Cs}$  and  $^{239,140}\text{Pu}$  from MAYAK present downstreams Dam 11 are found in the Techa river and floodplains. In case of  $^{90}\text{Sr}$  perhaps half of the MAYAK release may be found beyond the Techa, but the inventory estimate of  $^{90}\text{Sr}$  are encumbered with large uncertainties. It should furthermore be taken into account that in regularly flooded areas especially  $^{90}\text{Sr}$  may have penetrated deeper than 1 m (NREG, 1997).

In 1997 samples were collected further downstreams in the Iset and Tobol rivers. Table 5.4. shows some of the data.

Table 5.4. Cs-137 and <sup>239,240</sup>Pu in flooded areas along the Iset and Tobol (Samples collected July 1997)

River	Distance from MAYAK in km	kBq <sup>137</sup> Cs m <sup>-2</sup> 0-40 cm	<sup>239,240</sup> Pu/ <sup>137</sup> Cs	Sample type
Iset	310	23.6	0.030	foodplain soil
Iset	423	6.3	0.044	sorlake sediments
Iset	543	4.6	0.024	sorlake sediments
Tobol	723*/	7.6	0.0033	sorlake sediments
Tobol	778	2.9	0.0125	floodplain soil

\*/ The sample from Tobol (723 km) showed an enhanced <sup>137</sup>Cs level indicating an unknown source at the tributary Tura to Tobol river. The <sup>239,240</sup>Pu/<sup>137</sup>Cs was order of magnitude less than seen in the other samples.

Cs-137 in these samples were measured both by IPAE and RISØ. The mean ratio IPAE/RISØ was 1.37±0.66 (±ISD, N=73).

The range was 0.66-6.40. Low concentrations were in general overestimated by IPAE.

### 3.5.3 Collective doses to the world population following historical releases from Mayak PA

Detailed assessments of the transport of radionuclides from Mayak PA to the Kara Sea are all but absent in the open literature. In addition, the modelling of such processes in the Ob river system, has not featured strongly in the SUCON project. Nevertheless, some estimates of radionuclide input to the Arctic exist. Russian studies cited in Bradley & Jenquin (1995b) estimated that 0.17 PBq (4 700 Ci) of <sup>90</sup>Sr were carried to the Ob River and subsequently to the Kara Sea between 1961 and 1990 and the study of Trapeznikov *et al.* (1993) calculated that at least 1 PBq <sup>90</sup>Sr and <sup>137</sup>Cs was to be found in the Ob river system and Arctic Ocean.

In the SUCON project, dating of river sediment cores near Majak have been performed.

The cores were cut in slices (2 cm thickness) and analysed for <sup>210</sup>Po. The results show that only the profile from Reservoir 11 show a depth distribution of <sup>210</sup>Po that possibly could lead to a successful dating. The sedimentation rate for this core was determined to 3.1 mm/year.

The distribution of <sup>137</sup>Cs and <sup>90</sup>Sr in river water and sediments as a function of distance from dam 11 has also been studied. (Figs. 5.5. and 5.6).

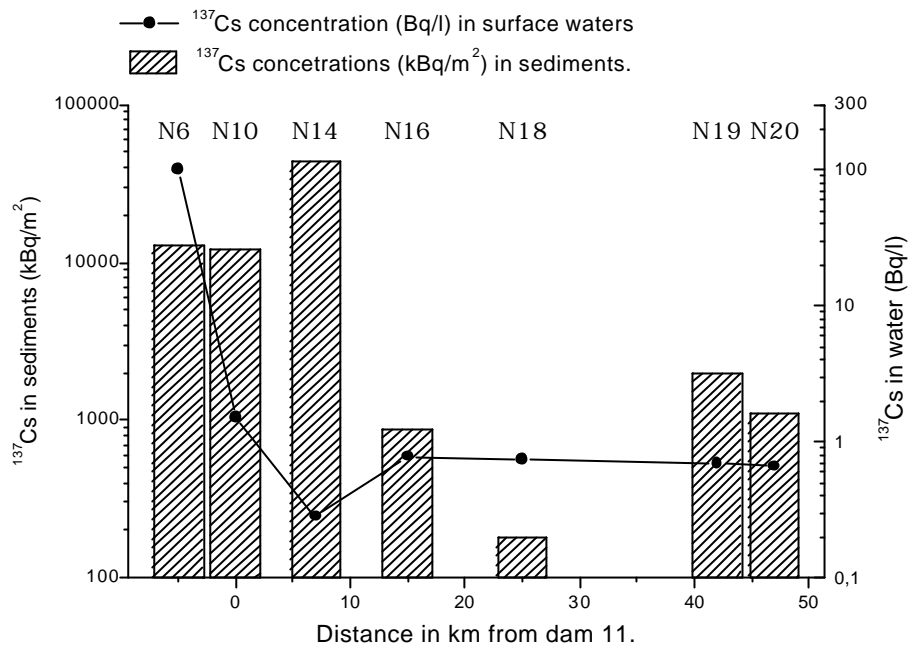


Figure 5.5. <sup>137</sup>Cs in sediments (kBq/m<sup>2</sup>) and water (Bq/l) downstream Techa River from reservoir 11 (JNRC, 1997).

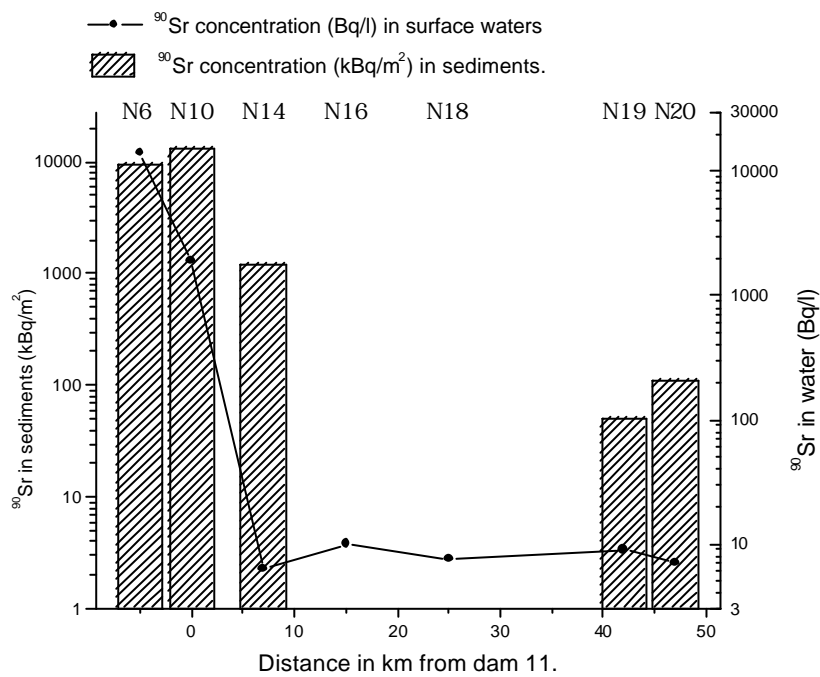


Figure 5.6. <sup>90</sup>Sr in sediments (kBq/m<sup>2</sup>) and water (Bq/l) downstream Techa River from reservoir 11 (JNRC, 1997).

The concentration of both <sup>137</sup>Cs and <sup>90</sup>Sr in water is very high in the reservoirs, and decreases significantly (factor of 10<sup>2</sup> for <sup>137</sup>Cs and 10<sup>3</sup> for <sup>90</sup>Sr) downstream Techa River from Dam 11 to Muslyumovo, 47 km from the dam. In Reservoirs as well as in the Asanov Swamp the very high levels of <sup>137</sup>Cs and <sup>90</sup>Sr are retained in the sediments. The contamination densities are significantly reduced (factor of 40 for <sup>137</sup>Cs, factor of 10 for <sup>90</sup>Sr) downstream Asanov Swamp.

In the absence of detailed quantitative information in relation to the transport of radionuclides from Mayak PA to the Arctic, the global impact, in terms of collective doses, of unit inputs (1 TBq) of radionuclides to Obskaya Guba (Ob Estuary) have been assessed. An input of this magnitude may be an underestimate for  $^{90}\text{Sr}$  over the history of Mayak PA operations but can be considered a reasonable, if not conservative, estimate for the other radionuclides considered ( $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ ) which either have relatively high  $K_d$ s in freshwater environments and/or have been discharged to the Techa River in much smaller quantities (as is the case for  $^{60}\text{Co}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Am}$ ).

Two models have been applied to derive collective doses to man arising from 1 TBq inputs of radionuclides to Obskaya Guba, namely a traditional box model (Nielsen *et al.*, 1997) and a derivative of this model which includes a module to account for the dispersion of radionuclides over time (Iosjpe & Strand, 1998). Results from the latter of these models is shown in Figure 5.7. which illustrates the spatial distribution in Arctic waters 4 years after a 1 TBq release of  $^{239}\text{Pu}$  to Obskaya Guba.

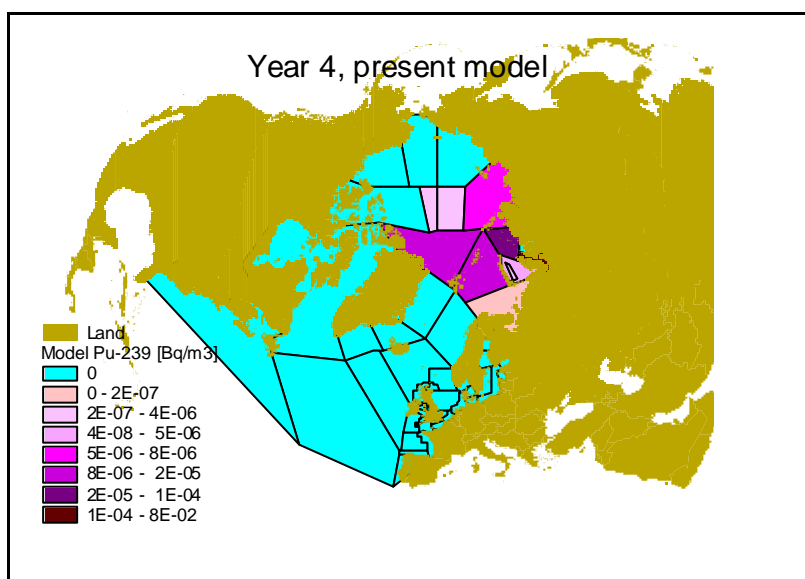


Figure 5.7. Specific activity level ( $\text{Bq m}^{-3}$ ) of  $^{239}\text{Pu}$  in water 4 years after a 1 TBq release of  $^{239}\text{Pu}$  to Obskaya Guba.

Calculated collective doses to man for a 1000 year period from a unit release are presented in Table 5.5. Model calculations are similar apart from the collective doses for  $^{241}\text{Am}$  and  $^{239}\text{Pu}$  which dominate doses in both model outputs. Highest doses are 0.03-0.22 manSv from  $^{239}\text{Pu}$  and 0.0029-0.06 manSv for  $^{241}\text{Am}$ . Assuming that 1 Pq  $^{90}\text{Sr}$  actually reached Obskaya Guba from Mayak PA in the early fifties the global collective dose from this contamination would have been 0.5 man Sv. This compares to a collective dose to Arctic populations arising from fallout from nuclear weapons testing of 13 000 manSv over the next 50 years (Strand *et al.*, 1997). Assuming the input scenario is a reasonable one, it appears that the global impact, in terms of doses to man have been and will be relatively low.

Table 5.5. World collective dose for 1000 years (manSv) following a 1 TBq discharge of radionuclides to Obskaya Guba.

Radionuclides	Iosjpe & Strand, 1998	Nielsen <i>et al.</i> , 1997
<sup>60</sup> Co	$1.5 \times 10^{-4}$	$2.1 \times 10^{-4}$
<sup>137</sup> Cs	$3.0 \times 10^{-3}$	$3.9 \times 10^{-3}$
<sup>90</sup> Sr	$4.7 \times 10^{-4}$	$5.7 \times 10^{-4}$
<sup>241</sup> Am	$2.9 \times 10^{-3}$	$6.1 \times 10^{-2}$
<sup>239</sup> Pu	$3.0 \times 10^{-2}$	$2.2 \times 10^{-1}$

## 3.6 WP 6 Speciation and Long Term Behaviour

### 3.6.1 Radionuclide species in water

Radionuclides in water and heterogeneous systems, sediment-water or soil-water systems, may distribute between low molecular weight forms (LMW), high molecular weight forms (HMW) and colloids in soil water, or are reversibly or irreversibly bound to particles (Fig 6.1). Thus, the distribution of radionuclide species will change with time due to particle weathering and interactions with naturally occurring components (e.g. humic substances, clay material).

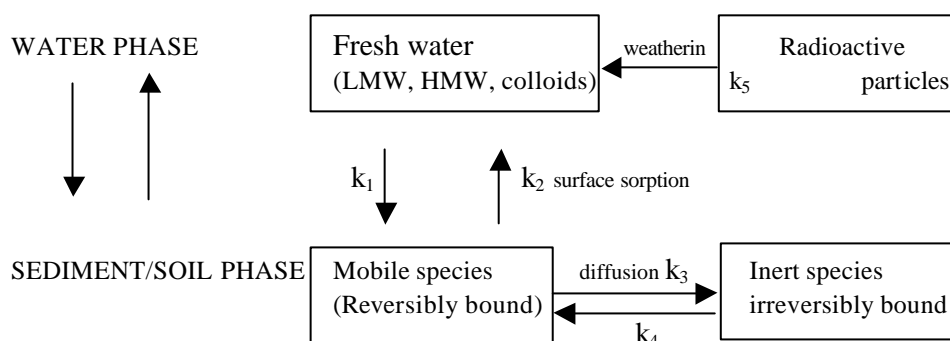


Figure 6.1. Distribution of radionuclide species in water, sediments-water and soil-water systems.

As the reservoirs are relatively shallow and exposed to wind erosion, the fraction of resuspended material ( $>0.45 \mu\text{m}$ ) was relatively large. Based on filtration ( $0.45 \mu\text{m}$ ) and hollow fibre ultrafiltration (10 kDa) about 10 - 50% of <sup>137</sup>Cs was associated with particles ( $>0.45 \mu\text{m}$ ) in reservoir and river water, while the colloidal fraction ( $10 \text{ kDa} < \text{size fraction} < 0.45 \mu\text{m}$ ) was insignificant. Thus, 50-90% of <sup>137</sup>Cs was present as LMW species in the water. For <sup>90</sup>Sr, LMW species ( $< 10 \text{ kDa}$ ) was predominantly present in the water. For Pu, a variable fraction was associated with suspended material.

### 3.6.2 Radioactive particles in soils and sediments

Information on physico-chemical forms, binding mechanisms and degree of binding to sediments is essential for transport modelling, as soil and sediment can act as a sink for strongly (irreversibly) bound radionuclides. Furthermore

contaminated sediments may act as a diffuse source for reversibly associated species which are remobilized in the future. In soils and sediments, especially in samples collected in the reservoirs and the Asanov flood plain, the radionuclides were inhomogeneously distributed. As localised heterogeneities indicate the presence of radioactive particles, autoradiography, synchrotron based X-ray absorption spectroscopy, grain size separation and extractions have been carried out.

Autoradiography (Fig 6.2) demonstrated the presence of localised heterogeneities. Furthermore, results from electron microscopy with X-ray microanalysis and synchrotron based microscopic X-ray absorption spectroscopy (XAS) documented the presence of large (>10µm) particles containing radionuclides as well as heavy metals (Salbu 1999).

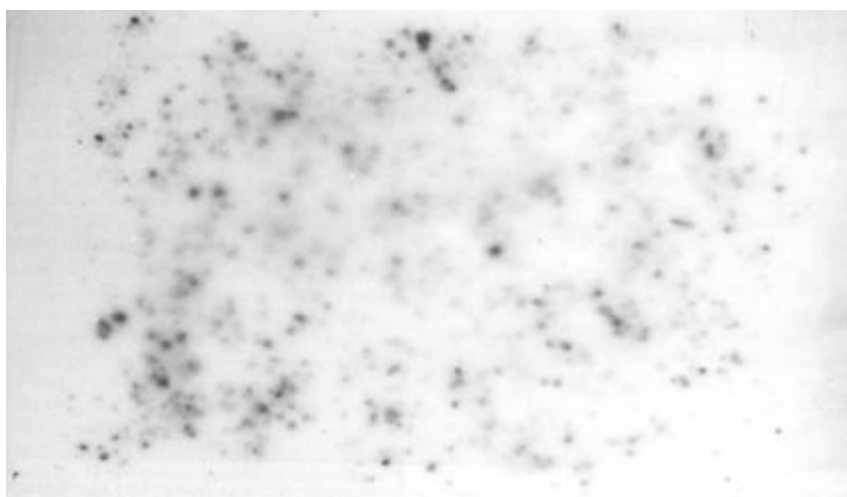


Figure 6.2. Autoradiography of sand sample from Reservoir 10.

Using wet sedimentation techniques, grain size analysis of sediments (Reservoir 10 and 11) and soils along the flood plain of River Techa downstream from dam 11 demonstrated in general that sand (30-88%) and silt (10-60%) were predominant (Table 6.1). The clay fraction amounted to less than 6%. Even though the clay content was small, clay in soil acted as an efficient sink for <sup>137</sup>Cs. Sequential extractions demonstrated that <sup>137</sup>Cs was irreversibly associated with components in the sand and silt fractions.

Table 6.1. Relative Grain size Distribution of <sup>137</sup>Cs activity in sediment and soil samples 1996.

km from dam 11	↓	<sup>137</sup> Cs		Sand		Silt		Clay	
		Bq/kg total	Activity (% tot)	Grain size (% tot)	Activity (% tot)	Grain size (% tot)	Activity (% tot)	Grain size (% tot)	
Reservoir 10, sediment	-	283000	84	81	15,7	19	0,3	0,4	
Reservoir 11, sediment	-	19000	49,9	32	45,1	61	5	6,7	
Down Techa river, soil	0	50000	66,3	71	32,3	28	1,4	1,6	
Down Techa river, soil	7	150	75,4	81	24,6	18	0	1	
Down Techa river, soil	80	1700	41,5	67	48,3	29	10,1	4,8	
Down Techa river, soil	245	180	3,2	88	16,7	10	80,1	1,2	

The data in table 6.1 show that there is a dramatic decrease in the total amount of Cs activity associated with soils and sediments, with distance from the reservoirs. Also, there is a change in the degree of association with the various fractions. Closer to the reservoirs, the bulk of the activity is associated with the sand and silt fractions, but further downstream, the clay fraction becomes more important, even though the relative proportions of each are fairly constant.

### 3.6.3 Mobility of radionuclides in soils and sediments

Radionuclides reversible or irreversible associated with solid phases in soils and sediments are studied by using sequential extractions (Oughton and Salbu 1994). The extractants are chosen to obtain information on:

- a) reversible physical sorption (inert electrolytes; water or 1 M NH<sub>4</sub>Ac at soil or sediment pH)
- b) reversible electrochemical sorption (pH effect; 1 M NH<sub>4</sub>Ac at pH 5)
- c) irreversible chemisorption (red/ox systems; hydroxylamin, H<sub>2</sub>O<sub>2</sub> in 1 M HNO<sub>3</sub>, 7 M HNO<sub>3</sub>)

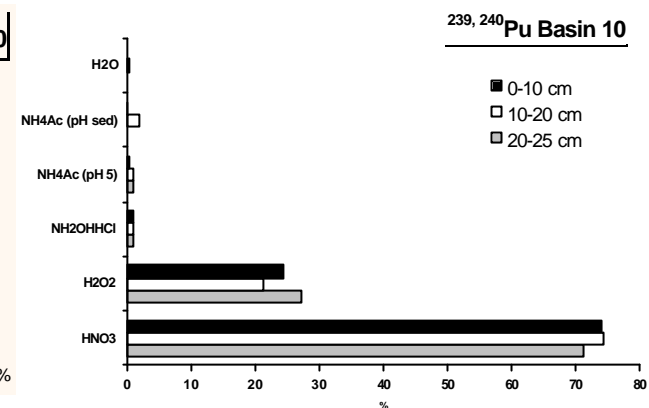
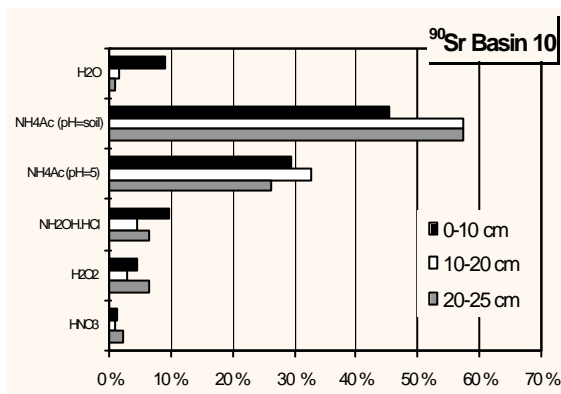
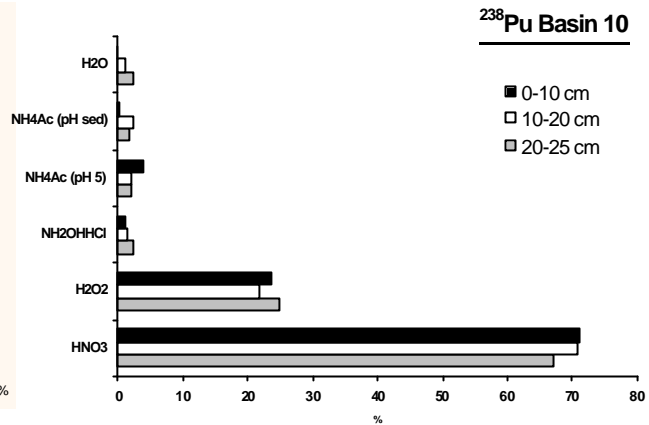
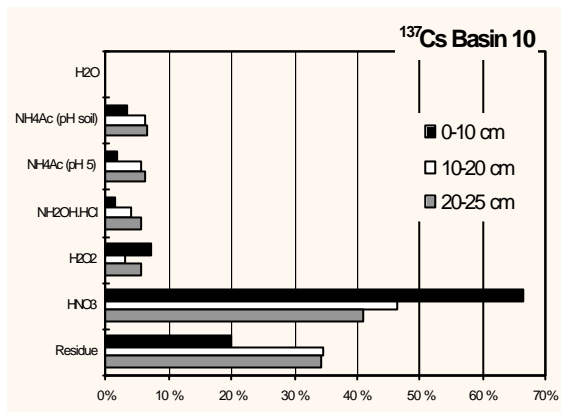
Radionuclide species reversibly bound are assumed to be potentially mobile, while species irreversibly bound to components in soil or sediments are considered inert.

The mobile fraction of radionuclides (i.e. H<sub>2</sub>O, NH<sub>4</sub>Ac extractable) can be clearly distinguished from inert forms being released after dissolution (breakage of chemical bonds due to red/ox agents). As reversibility is a matter of kinetics, the time needed for reversible processes (e.g. displacement reactions; physical or electrochemical sorption) to reach equilibrium may be considerable. Thus, the chosen contact time allows rapid surface processes to occur, while less attention is paid to the slow diffusion process. By comparing the extraction yields of radioactive and stable isotopes of Cs and Sr, information on particle association and the degree of isotopic exchange can be obtained.

In general, the mobile fraction (inert electrolyte or change in pH) of <sup>137</sup>Cs and Pu isotopes in soils and sediments are very low, less than 10% (Fig. 6.3) while a substantial fraction of <sup>90</sup>Sr is mobile. In general <sup>90</sup>Sr and stable Sr usually showed a rather similar distribution between extracted fractions. Thus, isotopic exchange seems to have reached pseudo-equilibrium. However, a certain fraction of <sup>90</sup>Sr is still rather strongly attached to soils and sediments. <sup>137</sup>Cs and Pu isotopes in sediments and soils is usually irreversibly sorbed (fixation or chemisorption) or incorporated in the mineral lattice. Thus, bonding rupture is needed for the species to be released and the most effective extracting agent for <sup>137</sup>Cs, stable Cs as well as Pu-isotopes in soil or sediment is 7 M HNO<sub>3</sub>. Variations in the relative distribution between HNO<sub>3</sub> and residual fractions in soils and sediments, can reflect different contact times (e.g. time dependent diffusion of Cs ions into the clay mineral lattice) and differences in the characteristics of soils and sediments. In most cases however, <sup>137</sup>Cs distribution between extraction fractions showed a similar distribution to that of naturally occurring stable Cs. Thus, 40-50 years after the major discharges of <sup>137</sup>Cs, the isotopic exchange seems also to be close to equilibrium.

Figure 6.3 Relative distribution of <sup>137</sup>Cs, <sup>90</sup>Sr and Pu-isotopes in sequential extraction studies from reservoir 10. The precision is within 10%.

Plutonium-isotopes are strongly associated with soils and sediments, however, more easily extracted with HNO<sub>3</sub> than <sup>137</sup>Cs. This could be explained by the fact that Pu-ions are sensitive to scavenger effects, e.g. coprecipitation with macro elements (e.g., Fe). Furthermore, an association with organic compounds is also indicated. In the reservoir sediments, <sup>238</sup>Pu and <sup>239,240</sup>Pu showed similar distributions between the extracted fractions (Fig. 6.3) throughout the profile (0-25 cm).



For radionuclides associated with uranium fuel particles, the particles are oxidised by  $H_2O_2$  in 1 M  $HNO_3$  and associated radionuclides are released (Oughton et. al 1993, Salbu et al. 1994 and 1998, Kashparov et. al, 1997). As  $^{90}Sr$  and stable Sr usually are mobile and  $^{137}Cs$  and stable Cs usually are extracted by 7M  $HNO_3$ , the presence of  $^{90}Sr$  and  $^{137}Cs$  in  $H_2O_2$  fraction indicates an association with radioactive particles. In the reservoirs, an increased yield of  $^{90}Sr$  (20 % in Reservoir 11),  $^{137}Cs$  (40-50% in Reservoir 10) and Pu-isotopes (20%) in the  $H_2O_2$  -  $HNO_3$  fractions may therefore reflect the presence of radioactive particles in sediments (Fig. 6.3 and 6.4a). As particle weathering occur,  $^{90}Sr$  is believed to be mobilised, while released  $^{137}Cs$  and Pu-isotopes probably will associate strongly with components in the sediment. Reservoir 10 substantial amount of U, lanthanoides and heavy metals were also present in the sediments collected close to the outlet of Reservoir 4.

The present results show that contaminated sediments and soils will continue to act as a source of mobile  $^{90}Sr$  to waters and biota. The activity levels in sediments and soils should however, be reduced due to long term wash-out of  $^{90}Sr$ . In contrast,  $^{137}Cs$  (and Pu) are strongly bound to sediments and soils and a transfer of  $^{137}Cs$  (and Pu) from sediments and soils to water should attributed to re-suspension of contaminated colloids or particles. This supports the suggestion that the low  $^{90}Sr/^{137}Cs$  ratio in Techa River sediments and the Asanov Swamp, as compared to Reservoirs 10 and 11, is due to the preferential remobilization of  $^{90}Sr$  from soils and sediments.



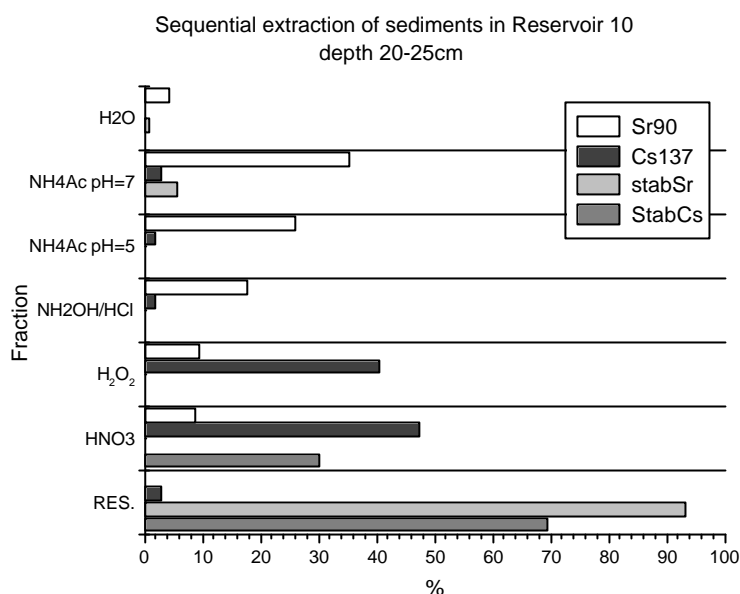


Figure 6.4a. Relative distribution (%) of  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ , stable Cs and stable Sr in sequentially extracted fractions of sediments from reservoir 10, (1996 field work). The precision is within 10%.

Another extraction experiment including four sediment samples from Reservoir 11 subjected to three extraction agents were performed to compare the possible differences in mobility between radioactive and stable Cs. The experimental work included the use of two weak extraction agents (1 N ammonium acetate and 1N ammonium nitrate) and one strong extraction agent (7N hot nitric acid) with the extractions performed in parallel on three aliquots of each sample. The content of radiocesium (i.e.  $^{137}\text{Cs}$ ) was determined by high resolution gamma spectrometry. Stable cesium ( $^{133}\text{Cs}$ ) was determined either by ICP-MS (extracts) or INAA (total content). The results (Fig. 6.4.6.) show significantly higher extractability for  $^{137}\text{Cs}$  than for stable Cs, indicating a chemical disequilibrium between these two isotopes with respect to mobility. This is probably what should be expected from the differences in physio-chemical characteristics between these two isotopes; stable Cs as an original part of the mineral structure whereas  $^{137}\text{Cs}$  has either been externally applied on the sediment particle surfaces or is contained in particles originating from the Mayak enterprise.

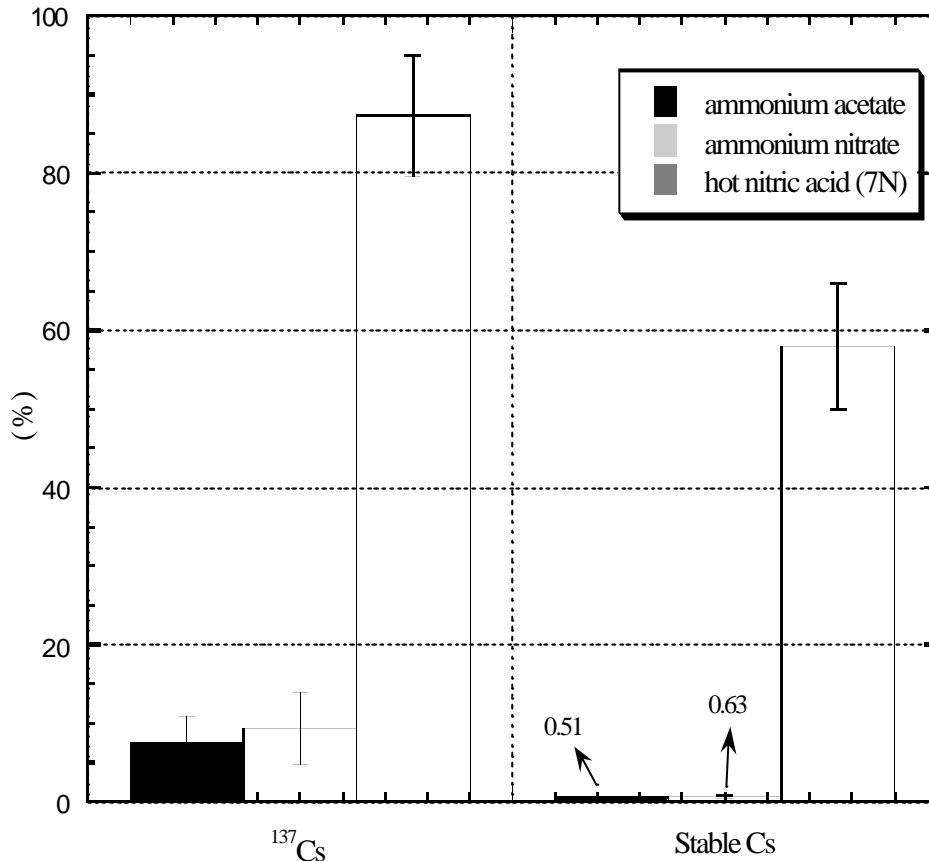


Figure 6.4b. Extractability of  $^{137}\text{Cs}$  and stable Cs relative to total content in sediment samples from Reservoirs 10 and 11. Error bars are given as standard error of the mean ( $n = 4$ ).

### 3.6.4 Interactions of different Pu-species

Information on the source term is usually restricted to the list of inventory and the activity levels. However, the physico-chemical forms of released radionuclides are essential for the interaction with soil components and the degree of which soils will act as a permanent sink or if contaminated soils can act as a diffuse source in the future. For multivalent elements like plutonium, releases from nuclear reactors or waste disposal sites may include Pu associated with fuel particles, Pu-colloids, oxidised forms ( $\text{Pu}^{\text{V,VI}}$ ), reduced forms ( $\text{Pu}^{\text{III,IV}}$ ) or organic complexes due to interaction of Pu-species with fulvic or humic materials.

To model the radionuclide transport in soil-water system, the equilibrium distribution coefficient  $K_d$  (Bq/kg soil d.w /Bq/l water) is usually obtained from samples collected in the field. If particles are present  $K_d$  will be overestimated. Furthermore, assumption of equilibrium conditions may not be valid. Thus, the time depended distribution coefficient have been studied by dynamic tracer experiments where different physico-chemical forms of Pu-isotopes have been added to Reservoir 10 sediment-water or Mayak soil-water systems. After different contact times the samples have been analysed and thereafter sequentially extracted to determine mobile and inert fractions (Skipperud et. al., in. prep.).

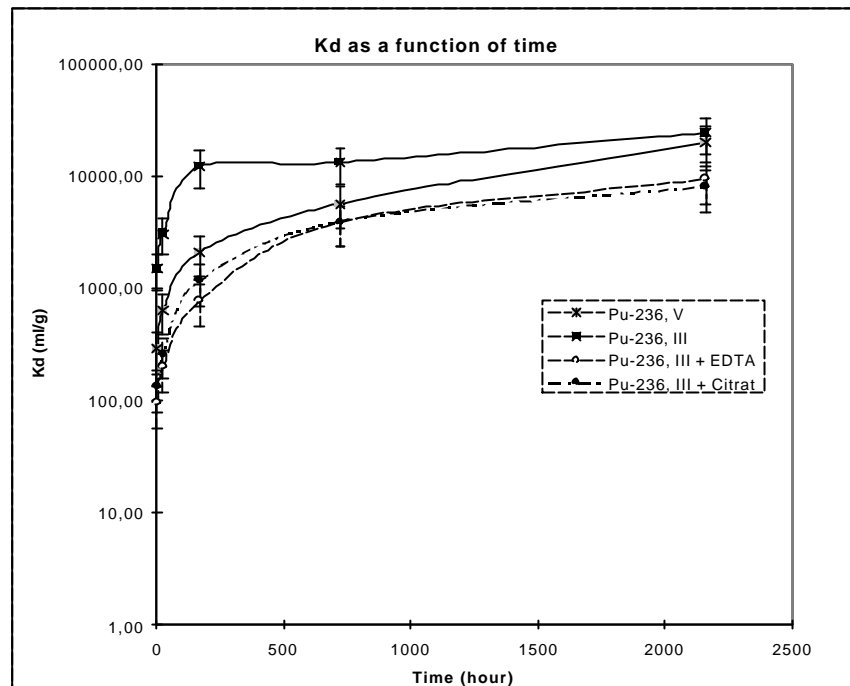


Figure 6.5. Distribution coefficient,  $K_d$ , of different physico-chemical forms of plutonium as a function of contact time ( $n=3$ ) (Skipperud et. al in.prep).

For the Pu-isotopes the rate of interactions as well as the  $K_d$  obtained after 2000 hrs contact time, was dependent on the Pu-specie in question. The interaction of Pu-species with soils is rapid and can be described by two first order equations  $k_D = ae^{-k_1t} + be^{-k_2t}$  (Fig. 6.5). The results shows that  $K_d$  of  $10^4$ - $10^5$  mg/g for Pu<sup>III,IV</sup> is about one order of magnitude higher than for Pu<sup>V,VI</sup> and Pu<sup>III,IV</sup>-organic after a very short contact time. Thus, Pu-species of interest and the time needed for equilibrium to be attained must be carefully considered when  $K_d$ 's are chosen for modelling purposes.

### 3.6.5 $^{240}\text{Pu}/^{239}\text{Pu}$ isotope ratio

The speciation of released radionuclides, especially multivalent nuclides like Pu, is usually source related. to identify contribution from different sources,  $^{240}\text{Pu}/^{239}\text{Pu}$  isotope ratios are determined by accelerator mass spectrometry (AMS). The lowest  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios, 0.014 - 0.024, were found at the Asanov Swamp, where the primary source of contamination was direct discharge of intermediate level radioactive waste between 1949 and 1951. The highest ratios, 0.06 - 0.29, were found in Reservoirs 10 and 11, which have been contaminated by various sources of effluents up to the present day (Fig. 6.6). Thus, the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio released from Mayak seems to have increased from 1949/51 until present.

The increase in the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio is a consequence of a decline and ventual halt in the production of weapons' grade Pu and an increasing focus on civil reprocessing at the plant since the 1960's. Furthermore, measurement of Pu isotopes in low-level samples collected from the Techa, Iset and Ob Rivers showed that while activity levels decrease with distance from Mayak - from 2000 Bq/kg at 7 km downstream to less than 1 Bq/g sediment at 250 km, the  $^{240}\text{Pu}/^{239}\text{Pu}$  isotope ratios increase. These results suggest that most of the plutonium in the Upper Techa River originates from early waste discharges. However, enhanced atom ratios in surface sediments at Muslyomovo and downstream (0.035 -

0.099) could indicate an influence of additional, later sources. At greater distances from the plant, the influence of global weapons fallout becomes progressively more apparent as isotope ratios increase. In particular, surface sediments collected from the Ob Estuary showed activity levels and Pu-isotope ratios consistent with global weapons fallout.

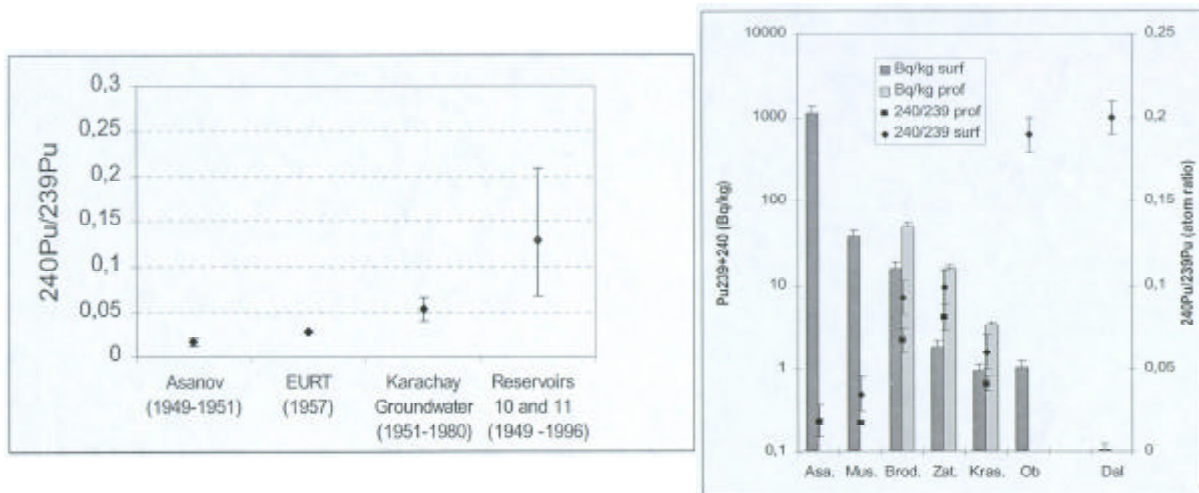


Figure 6.6.  $^{240}\text{Pu}/^{239}\text{Pu}$  isotope ratios for Asanov Swamp, EURT, Lake Karachay and Reservoir 10 and 11 and  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$  activity concentrations (Bq/kg) and  $^{240}\text{Pu}/^{239}\text{Pu}$  isotope ratios in surface sediments and sediment profiles in the Techa, Iset and Ob Rivers (Oughton et. al. In prep).

### 3.6.6 Tc-99 studies in the Techa river

Sediments collected from the Techa River at Nadirov Bridge were analysed for  $^{99}\text{Tc}$ . The maximum concentration was found at a depth of 14-18 cm. This layer was sedimented around 1950, i.e. at a time when the maximum releases of waterborne radionuclides occurred to the river from the Mayak. The vertical distribution of  $^{99}\text{Tc}$  in sediments followed that of  $^{137}\text{Cs}$  (see Fig. 6.7) and the mean ratio between  $^{99}\text{Tc}$  and  $^{137}\text{Cs}$  in the sediments at the time of discharge was  $0.41 \times 10^{-3}$ . Laboratory experiments showed that  $^{99}\text{Tc}$  under anoxic conditions, e.g. when organic material is decomposing, will be retained by sediments.

If the  $^{99}\text{Tc}$  to  $^{137}\text{Cs}$  ratio found in the sediments is representative for the ratio in the liquid discharges from MAYAK, i.e. if  $^{99}\text{Tc}$  and  $^{137}\text{Cs}$  under the given circumstances had the same  $K_d$  the discharge of  $^{99}\text{Tc}$  is calculated to  $0.41 \cdot 10^{-3} \cdot 12.2 \text{ PBq} = 5 \text{ TBq } ^{99}\text{Tc}$ . However, this figure is probably a minimum estimate of the  $^{99}\text{Tc}$  discharge as it seems unlikely that all  $^{99}\text{Tc}$  should have been retained just as readily by the sediments as  $^{137}\text{Cs}$ . If the ratio in the discharge had been the same as in the discharges from Sellafield i.e.  $2.1 \cdot 10^{-2} \cdot 12.2 \text{ PBq} = 0.3 \text{ PBq } ^{99}\text{Tc}$  had been discharged and most of this would then have reached the Arctic Ocean through the Ob river system. If such an input of  $^{99}\text{Tc}$  had occurred to the Arctic Ocean around 1950 it should be possible to see a signal in brown algae (e.g. *Furoids*) which concentrate Tc from seawater by a factor of  $10^5$  if any seaweed, e.g. in herbarium, have been preserved from that time.

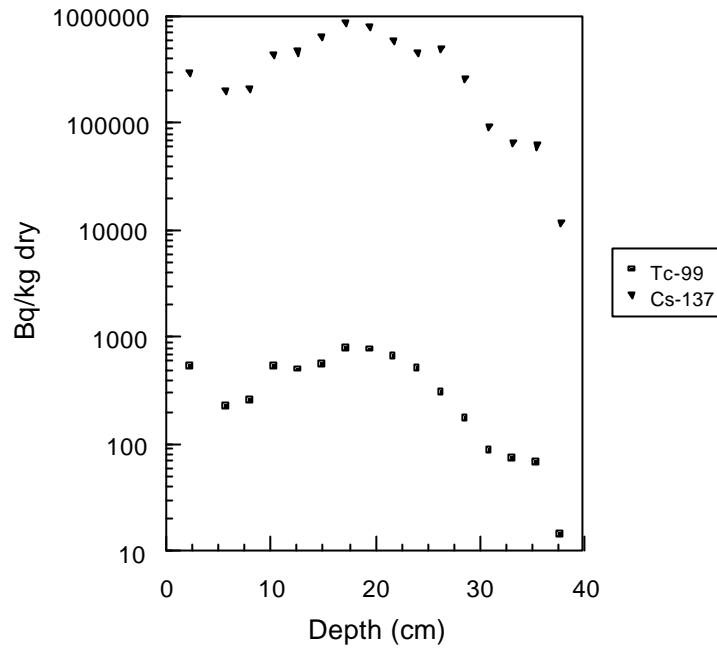


Figure 6.7. Depth distributions of  $^{99}\text{Tc}$  and  $^{137}\text{Cs}$  concentrations in Techa river sediments from Nadirov Bridge.

### 3.6.7 Studies of $^{237}\text{Np}$ and other transuranic elements in the Mayak drainage area.

Figures 6.8 and 6.9 show depth distributions of activity concentrations and atom ratios for plutonium and neptunium in a Techa River sediment core taken at Nadirov Bridge approximately 50 km downstream from the release point to the Techa River from the Mayak complex. Other radionuclide data from this core are given elsewhere Trapeznikov *et al* (1993) Aarkrog *et al* (1997). It is seen that although there is a considerable variation in concentrations with depth, the  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios are very stable with depth, or in other words, with time. The average  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratio show a remarkably low variation of the same size as the average statistical error of the ICPMS ratio measurements,  $0,0167 \pm 0,0005$  (SD, n=14).

For the  $^{237}\text{Np}/^{239}\text{Pu}$  atom ratios (Fig. 6.9), the data indicates a peak at 20-26 cm. The significance of this excursion has not yet been verified by repeated measurements. It should be born in mind, that the  $^{237}\text{Np}/^{239}\text{Pu}$  ratio is determined with a much higher uncertainty than the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio because the amount of  $^{237}\text{Np}$  is determined on the assumption of a predetermined average chemical yield and furthermore, the  $^{237}\text{Np}/^{239}\text{Pu}$  ratio is based on two individual chemical separations and two ICPMS measurements, whereas the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratio is determined from one ICPMS run Stürup *et al* (1998).

The  $^{237}\text{Np}/^{239}\text{Pu}$  ratios from the Nadirov Bridge core (Fig. 6.9) are plotted as a function of the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios in figure 6.10, together with other data from the Techa River, the Ob River, and the Iset River. As compared with Beasley *et al.*'s (1998) relation between the two atom ratios in Northern Hemisphere soils, shown as a line in Fig. 6.10, the Techa River samples show lower  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios. The Ob River and the Iset River data appears to have less  $^{237}\text{Np}$  compared to the  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios. Different sources or differences in geochemical properties of neptunium and plutonium could cause this. However, bearing in mind the above remarks on uncertainty of the  $^{237}\text{Np}/^{239}\text{Pu}$  ratios, the difference

from Beasley *et al.*'s line is probably not significant on the present data except for the lower  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios in the Techa River sediments.

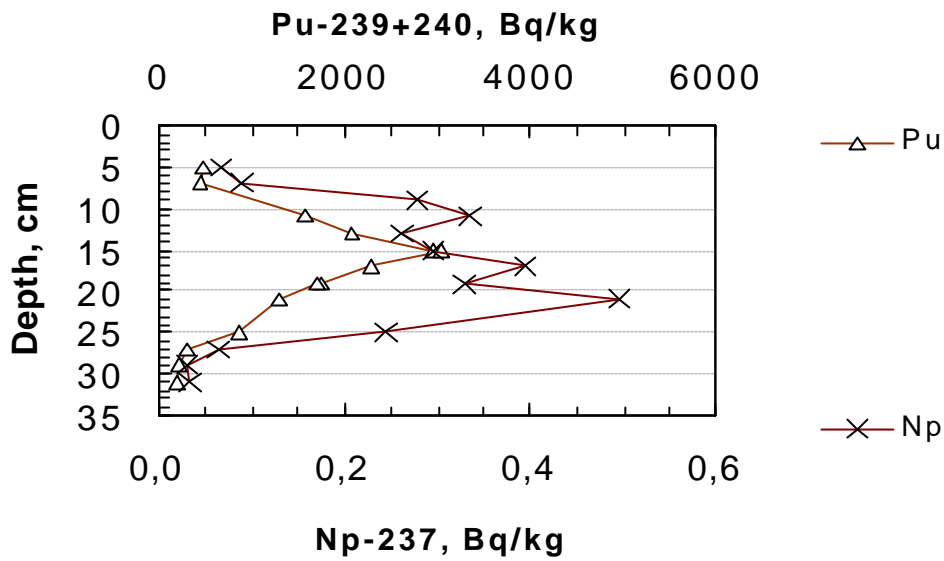


Figure 6.8. Nadirov Bridge, Techa River 50 km from Mayak. Depth distribution of  $^{239+240}\text{Pu}$  and  $^{237}\text{Np}$  activity concentrations.

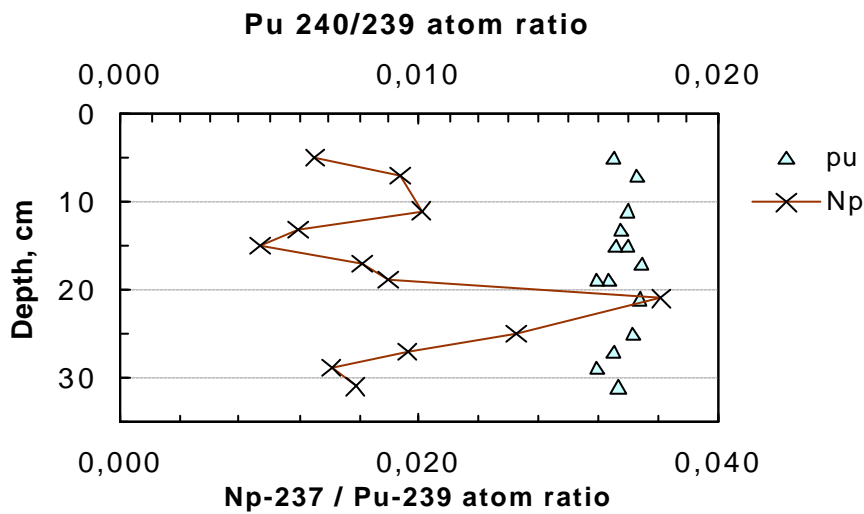


Figure 6.9. Nadirov Bridge, Techa River 50 km from Mayak. Depth distribution of  $^{240}\text{Pu}/^{239}\text{Pu}$  and  $^{237}\text{Np}/^{239}\text{Pu}$  atom ratios.

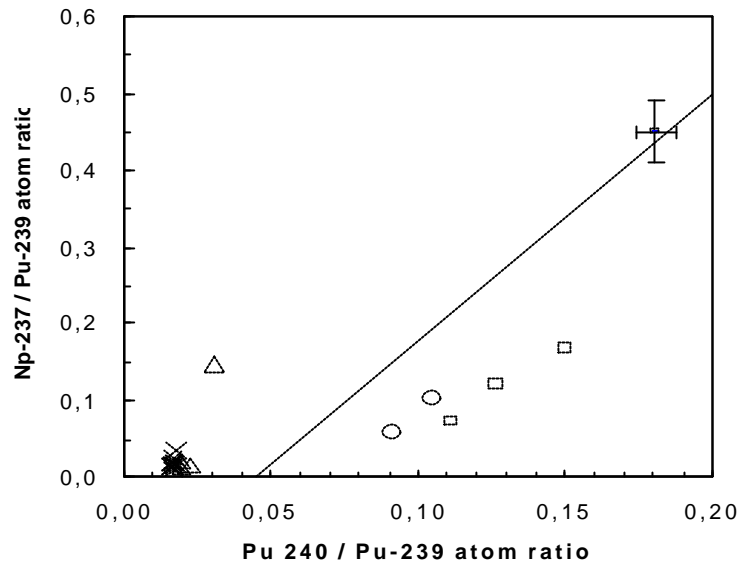


Figure 6.10.  $^{237}\text{Np}/^{239}\text{Pu}$  versus  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios in various Urals flood plain and river sediment samples.  $\times$ : Nadirov Bridge (Fig 1),  $\Delta$ : Anchugova, Techa 170 km from Mayak,  $\square$ : Ob River,  $\circ$ : Verkney Jahr, Iset River before confluence with Techa River. The line is Beasley et al.'s (1998) relation for northern hemisphere soils, and the single point indicate integrated fallout in Northern Tier Soils with 1  $\sigma$  errors.

### 3.6.8 I-129 in Techa river sediments

Sediments collected in 1992 at Nadirov Bridge (see 3.6.6.) were analysed for  $^{129}\text{I}$  by neutronactivation analysis. Fig. 6.11 show the results, which are comparable with those obtained in the neighborhood of other nuclear reprocessing plants. The ratio between  $^{129}\text{I}$  and  $^{137}\text{Cs}$  in the sediment column shows a minimum around 20 cm depth where the ratio is nearly an order of magnitude less than in the top and the bottom of the column. The high ratio in the deeper parts of the sediments may be due to a more rapid movement of the anionic iodine than of the cationic caesium.

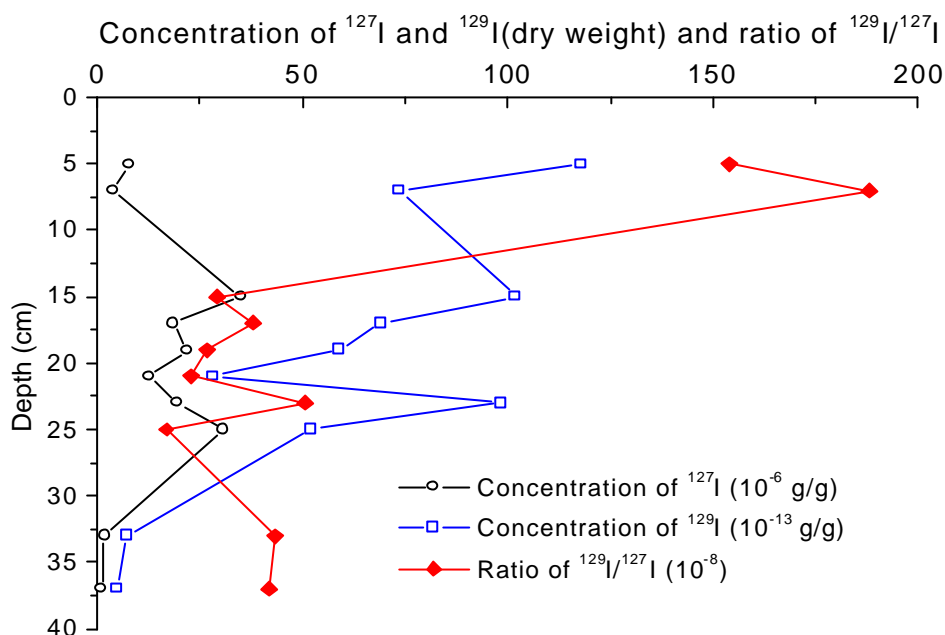


Figure 6.11. Depth distribution of  $^{129}\text{I}$  and  $^{127}\text{I}$  concentration and  $^{129}\text{I}/^{127}\text{I}$  ratio in sediment collected from Nordirov Bridge, Techa River, Russia

## 4 Main Achievements

A major achievement of the SUCON project has been the establishment and development for a fruitful co-operation between radioecologists from the Former Soviet Union and Western Europe.

Such collaboration has, for example, led to compilation of a Russian-Norwegian report (JNREG, 1997) which comprehensively deals with subjects ranging from the operation of the Mayak Production Association in terms of the handling of radioactive waste through to historical sources of radioactivity and present-day levels of contamination of the environment. This information has been invaluable input to the SUCON project, allowing further assessments of the levels of contamination to be undertaken and the identification of areas where further studies were urgently required.

The Russian-Norwegian report has shown:

Asanow Swamp is today the main source of strontium-90 to the Techa River. The amount of strontium-90 entering the Techa River below the swamp is decreasing but is estimated to be 1-2 TBq/year.

The level (1993-data) of radioactivity in Lake Karachay is 4.4 EBq, of which 40% are  $^{90}\text{Sr}$  and  $^{90}\text{Y}$  and 60%  $^{137}\text{Cs}$ . 98% of the activity contained in reservoirs at Mayak is located in Lake Karachay.

The early estimates of plutonium releases to the environment is too low. Official Russian data of plutonium releases is 2 TBq. Estimates of present levels of plutonium only in Reservoir 10 and 11 (1993-data) is 46 TBq.

Except for the plutonium inventory, the field work results from the joint project generally confirm the official Russian information.

Groundwater contamination from Lake Karachay mainly consists of strontium-90 and cobalt-60. The groundwater contamination is today of great con-



cern and are threatening the drinking water supply for the Novogornyi Village, a few kilometres south of Lake Karachay. Radionuclides from Lake Karachay has not yet reached the Techa River through the right bank channel (Mishelyak River).

Field measurements in the East Ural Radioactive Trace (EURT) due to the Kyshtym accident in 1957, but also contaminated from the Karachay wind dispersion in 1967, have shown:

- The deposition of  $^{137}\text{Cs}$  in the contaminated area may be an order of magnitude higher than that
- expected from the Kyshtym and Karachay accidents.

The inventory of  $^{90}\text{Sr}$  outside the near zone of Mayak calculated from soil samples is compatible with the Russian estimates.

The depositions of  $^{239,240}\text{Pu}$  outside the near zone from the Kyshtym and Karachay accidents were both about 1 TBq or in total approximately 1 kg plutonium. The deposition of Pu has not previously been determined.

Inventory estimates based on recent Russian studies, which has been synthesised through Norwegian-Russian bilateral work (JNREG, 1997) are summarised in Table 4. The uncertainties associated with previous inventory estimates has clearly identified the need for further studies involving the determination of radionuclide contamination in environmental samples.

*Table 4. Total inventory of radionuclides (primarily  $^{137}\text{Cs}$  and  $^{90}\text{Sr}$ ) in vicinity of Mayak PA.*

Location	Lake Karachay	Reservoir 17	EURT	Karachay area	Reservoir 2	Reservoir 3 & 4	Reservoir 10 & 11	Asanov Swamp
<b>Inventory (PBq)</b>	4 400 <sup>a</sup>	74 <sup>a</sup>	0.75 <sup>b</sup>	0.0067 <sup>b</sup>	0.74 <sup>c</sup>	1.9 <sup>c</sup>	7.9 <sup>c</sup>	0.25 <sup>d</sup>

<sup>a</sup> = Activity levels in 1993; <sup>b</sup> = Decay corrected release to 1996 with some activity removed due to catchment runoff and deep burial; <sup>c</sup> = activity levels in 1994; <sup>d</sup> = activity levels in 1992.

An assessment of current and future doses has been made for people living at three different locations in the Southern Urals region of Russia. The Techa river is a significant source of radiation exposure for the people who live alongside it. However, for both settlements considered, Brodokalmak and Muslyumovo, radiation doses are relatively low and less than  $1 \text{ mSv y}^{-1}$ , provided that the restrictions in place are followed. The most important exposure pathways are the ingestion of radionuclides in freshwater fish, milk and domestic vegetables. As expected given its location closer to the MAYAK site, the estimated doses at Muslyumovo are higher than those at Brodokalmak. In both locations if the restrictions were removed completely, such that people drank water and ate fish from the river, had free access to the river bank and grazed animals there, a significant increase in radiation doses would result. The most important pathways in this case are ingestion of radionuclides in fish from the river and in milk from cows grazing on the river flood plains. For Brodokalmak a probabilistic assessment of doses was also carried out taking account of the uncertainties on the input parameters to the assessment. This confirmed that doses were within acceptable ranges if restrictions were maintained but not if the restrictions were removed. There is the potential for doses to increase significantly if there are increases in the releases to the Techa river from the MAYAK site due to severe flooding or a dam failure.

For people living outside the restricted area set up following the Kyshtym fire accident in 1957, for example in Bagyarak, radiation doses are relatively low and there is no indication that any remediation measures are required. One of the most important exposure pathways is the consumption of freshwater fish. It may be possible to reduce the restricted area but further measurements and a subsequent dose assessment would be required.

Possible remediation measures for the region have been considered and recommendations made. These recommendations were based on reviews carried out of the measures taken in the region and of remediation measures that have been carried out or considered elsewhere. In addition, the radiation dose assessment considered the impact of any remedial measures in place. No further countermeasures are required for the Techa river population, although the existing measures need to be maintained. There is a concern about potential future doses given the significant quantities of radionuclide within the MAYAK site. The main recommendation is the need to safeguard against possible increases in discharges to the Techa river and the establishment of some form of monitoring system to alert the population in the event of large increases in the radionuclide concentrations in the Techa river.

There are possible measures that could be taken to reduce radiation exposures in the restricted areas around the MAYAK plant. These include alternative land use, land improving soil treatments and phytoremediation.

The extensive studies carried out to assess the global impact in particular of the Mayak contamination of the Ob river system have shown:

- In the lower reaches of Ob the radionuclide levels in river sediments collected from surlakes show no indication of contamination which could be attributed to early radioactive discharges (around 1950) from Mayak. The major source is global fallout. However, isotopic analysis of Pu and other transuranic elements suggest a contamination of the Ob-sediments from other sources than global fallout and the Chernobyl accident. Probably from the Pu-production at Tomsk-7.
- In the Techa-Iset rivers sediments as well as floodplain soil contain clear signals of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$  and Pu, Am which can be attributed to discharges to the Techa river from Mayak. Until now this contamination has been measured out to a distance of 300 km from Mayak.

The longitudinal radioactive contamination of the Techa-Iset river system and floodplains is decreasing with the distance from the release point at MAYAK and may be modelled by a power function. The vertical distribution of the activity in floodplains and sediments are modelled by an exponential function and the horizontal distribution of the contamination of the floodplain also decreases exponentially with the distance from the riverbed. Based on these model assumptions the inventories of the Techa river floodplain and river sediments downstreams the reservoir system at MAYAK were calculated to 0.1 PBq  $^{90}\text{Sr}$ , 0.3 PBq  $^{137}\text{Cs}$  and 0.8 TBq  $^{239,240}\text{Pu}$ . The uncertainty of these estimates is a factor of 3-4. Whereas essentially all MAYAK derived  $^{137}\text{Cs}$  and  $^{239,240}\text{Pu}$  are to be found in the Techa river system, about half of the  $^{90}\text{Sr}$  may be present downstreams Techa, but this is tentative.

Age determinations of the sediment layers from surlakes suggest that the major part of the MAYAK contamination of downstream parts of the Techa and Iset rivers first occurred 10-15 years after the high releases took place. Whether this has been due to a flooding of the Asanov Swamp in the early sixties or to a da-failure in reservoir 11 has not been answered yet.

Samples of river and floodplain sediments along the Iset and Tobol rivers have shown the presence of an unknown source of contamination at the river Tura (a tributary of Tobol).

The global collective dose from the release from Mayak in the early fifties, which are assumed to have contaminated Obskaya Guba with 1 TBq  $^{239, 240}\text{Pu}$  and 1 PBq  $^{90}\text{Sr}$  has been calculated to 0.5-0.8 man Sv.

The speciation studies in the reservoirs and in the Techa river has shown:

- In water,  $^{90}\text{Sr}$  is present in low molecular weight mobile forms, while a certain fraction of  $^{137}\text{Cs}$  and Pu-isotopes are associated with suspended particles.
- At least in Reservoir 10, radioactive particles are present in the sediments.
- In sediments and soils,  $^{90}\text{Sr}$  is predominantly present in potentially mobile forms. Thus, contaminated sediment and soil will continue to act as a source of mobile  $^{90}\text{Sr}$  to water and biota.
- Sediments and soils act as a sink for  $^{137}\text{Cs}$  and Pu-isotopes. Transfer to the water phase is attributed to resuspension of contaminated colloids and particles.
- The interactions of Pu-isotopes with sediments and soils depend on the physico-chemical forms of Pu. The time dependant distribution coefficient for  $\text{Pu}^{\text{III,IV}}$ -ions in soils is a factor of 10 higher than for  $\text{Pu}^{\text{V,VI}}$ -ions and Pu-organic compounds.
- The  $^{240}\text{Pu}/^{239}\text{Pu}$  isotope ratios in releases from Mayak seems to increase from 1949/51 (Asanov) until present (Reservoir 10 and 11).
- The  $^{240}\text{Pu}/^{239}\text{Pu}$  isotope ratios increase downstream Techa River, and an additional source is indicated downstream from Muslyomovo.

In the study of less well known radionuclides the SUCON project has determined  $^{99}\text{Tc}$ ,  $^{129}\text{I}$  and  $^{237}\text{Np}$  and plutonium isotope ratios in a number of highly contaminated samples. To this purpose radiochemical methods have been developed.

$^{237}\text{Np}$  and  $^{129}\text{I}$  has for the first time been reported in Techa river sediment samples.

## 5 Discussion

The studies conducted within Work-Package 1 of the SUCON project, have illustrated the fact that uncertainties associated with inventory estimates exist. To this end, Geographical Information Systems (GIS) could be employed in order to integrate disperse information on radionuclide contamination, geomorphology etc. into a more structured spatial format. By these means new inventory maps could be created to complement the existing maps that have been presented in the present project. Another study area of interest is the uptake and transfer of  $^{90}\text{Sr}$  in aquatic ecosystems. Although some work has been conducted on this topic, under field conditions, following atmospheric atomic weapons testing and the Kyshtym accident, the high levels of contamination in the Techa River system provide us with a unique opportunity to develop our understanding of  $^{90}\text{Sr}$  biogeochemistry.

There are a number of areas where further data would be useful to improve the understanding of the radiological impact of the contamination in the Southern Urals region. These include:

- Obtaining additional data on the levels of radionuclides in fish obtained from different lakes in the region, particularly those where different population groups obtain their fish. This would improve the dose assessment and, if there are differences between the concentrations, identify the best sources of fish.
- The dose assessments could be further improved with additional measurements of radionuclide concentrations in terrestrial foods particularly for Musylumovo and Bagaryak. It would also be worth considering additional radionuclides such as plutonium-239 and americium-241.
- The possibility of people living along the Techa river hunting and eating wild fowl should be investigated and measurements of radionuclide concentrations in the birds obtained to estimate possible doses.

Although the SUCON project failed to show any tracers from MAYAK in the downstream parts of the Ob river system and in the Kara Sea, it is still believed that the early releases of in particular  $^{90}\text{Sr}$  reached the Arctic Ocean around the 1950. To get the final proof for this assumption continued efforts should be made to obtain museum specimens of biota collected in the early fifties in the Arctic waters.

The relative high environmental contamination with longlived radionuclides, such as  $^{99}\text{Tc}$ ,  $^{129}\text{I}$ ,  $^{237}\text{Np}$ ,  $^{239,240}\text{Pu}$  and other transuranic elements makes the South Ural a unique study area for the radioecology of these radionuclides. SUCON has demonstrated that it by advanced radioanalytical and mass spectrometric methods is possible to measure these radionuclides in abiotic samples such as soil and sediments. A further analytical development should be encouraged in order to reduce the detection limits so measurements of these radionuclides could be made also on biota. This would make it possible to determine e.g. transfer coefficients.

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Abstract (max. 2000 characters)

This report summarises work done on the SUCON Project during 1996-1999 (European Commission Contract No. FI4C-CT95-0001). The project has focused on three major objectives: 1) An assessment of the radiological consequences of the contamination of the South Urals and the Ob river system from the production of plutonium at "Mayak", 2) The development of models to calculate doses to individuals and populations in the South Urals using environmental data, and 3) The intercomparison, harmonisation and standardisation of techniques used in dose reconstruction and specification of good practice in particular with regard to remedial measures.

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